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AUTHOR(S): Brad Lee Holian

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THE INTERACTION BETWEEN CESIUM AND GRAPHITE
FOR USE IN THE STUDY OF SURFACE PHENOMENA*

Brad Lee Holian
University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico 87545

Abstract

Surface diffusion has been hypothesized as the fast mode of an unusual fast-slow, two-mode transport process that has been observed in recent diffusion experiments with cesium in graphite. An interaction potential between a cesium atom and a graphite surface is obtained in order to study this surface diffusion by computer simulation (molecular dynamics method). At low surface coverage, the interaction between cesium atoms can be ignored so that the motion of only one cesium atom need be followed, albeit in a very complicated potential energy surface. Cesium is spontaneously ionized by graphite, so that the interaction of cesium with the graphite surface contains pairwise $\text{Cs}^+ - \text{C}$ terms (valence, induction, and dispersion forces) as well as an image-charge model of the bulk electrostatic interaction. All parameters but the strength of the repulsive $\text{Cs}^+ - \text{C}$ force are obtained by theoretical estimates, while this last parameter is determined by requiring that the adsorption $\text{Cs}^+ - \text{C}$ bond length be the same as observed in cesium-graphite lamellar compounds. Results indicate that the adsorption energy for a pit in the graphite surface of one to five missing carbon atoms is not greatly increased over that for the perfect surface (the one-atom hole is slightly repulsive compared to the perfect surface). For the hexagonal six-atom pit, the adsorption energy increases dramatically from about 120 kcal/mole for the perfect surface to about 200 kcal/mole and remains essentially constant for larger holes. Preliminary dynamical results for a cesium ion on a perfect graphite surface show free particle motion at high temperatures, necessitating the presence of defects in the graphite surface for truly diffusive motion.

I. Introduction

Recent experiments on the diffusion of cesium in graphite have demonstrated unusual transport behavior which cannot be explained by simple Fick's Law diffusion (1). Two

*Work performed under the auspices of the U. S. Energy Research and Development Administration and the Nuclear Regulatory Commission (Reactor Safety Research Division).

transport modes have therefore been hypothesized: 1) slow bulk diffusion and 2) fast surface diffusion. One proposed model (2) for this complex behavior includes, in two coupled diffusion equations, a first-order reversible exchange process between diffusing species - one located on the free surfaces of macroscopic pores as well as on the relatively free surfaces between the microcrystallites of graphite ("micropores"), and another population located in the more restricted regions between crystallites ("grain boundaries"). The transport of the latter population corresponds to the slow so-called bulk diffusion, even though large atoms like cesium cannot really penetrate into the bulk of the grains by lamellar diffusion (by squeezing in between the graphite layers). The diffusion along free surfaces proceeds much more rapidly, although the distinction becomes less obvious as the separation of the surfaces narrows.

In order to study the mechanisms involved in surface transport, molecular dynamics computer simulations of the motion of a cesium atom across a graphite surface have been undertaken. The classical equations of motion need be solved for only one cesium atom, since the behavior at low surface coverage, where the interaction between adsorbed cesium atoms can be neglected, is of particular theoretical and practical interest. Even with this simplification of the dynamics to the motion of one particle, the force acting on the cesium atom is quite complicated. In this paper, structural considerations and theoretical estimates of atomic properties will be combined to give a reasonable potential energy function for use in subsequent molecular dynamics calculations. The following six sections describe individual terms in the potential, with results presented in the last section.

II. Zero of Energy

The zero of energy for a cesium atom interacting with the graphite surface will be taken to be that of the atom separated infinitely far from the graphite crystal. As the atom approaches the surface, the electronic configuration of the atom becomes more and more distorted. The outer 6s electron, being loosely bound to the core, is easily snatched from the cesium by the metal-like graphite surface plane. When an electron is donated to the conduction band of the graphite plane (chemical symbol of graphite, Gr) energy is released - the electron work function, $E_w(\text{Gr})$:



The first ionization of cesium requires energy - the ionization potential, $E_I^{(1)}(\text{Cs})$:



The overall reaction [the sum of Eqs. (1) and (2)] requires energy W:



where $W = E_I^{(1)}(\text{Cs}) - E_w(\text{Gr})$ is negative, that is, heat is given off by the reaction. In units of $\text{kK} = 1000 \text{ K}$, $E_I^{(1)}(\text{Cs}) = 45.18$, $E_w(\text{Gr}) = 53.62$, and $W = -8.44$. The values of W for the alkali metal series Li, Na, K, Rb, and Cs are, respectively, 8.93, 6.01, -3.27, -5.16, and -8.44 kK . Hence, K, Rb, and Cs form stable lamellar compounds with graphite, while Li and Na do not (3).

III. Bulk Electrostatic Interaction

The ionized cesium atom interacts with its donated electron in the graphite conduction band much like a point charge over an infinite perfectly-conducting plane - the classical electrostatic problem of the image charge. The principal uncertainty in this model is the location of the conducting plane. If we assume that the cesium atom is centered over a hexagon of carbon atoms at a height of $z_m = 2.97 \text{ \AA}$ above the graphite plane [(as in the case of lamellar cesium-graphite compounds (3,4)], then the height of the conducting plane above the plane of the carbon nuclei z_o is simply $z_m - r(\text{Cs}^+)$, where $r(\text{Cs}^+) = 1.69 \text{ \AA}$ is the ionic radius of cesium (5). This is consistent (3) with a Van der Waals' radius for a graphite carbon atom of $r(\text{C}) = 1.602 \text{ \AA}$ (the C-C bond length in graphite is $r_o = 1.421 \text{ \AA}$ and the spacing between layers is $c_o = 3.354 \text{ \AA}$); $r(\text{C})$ is therefore very close to $1/2 c_o = 1.677 \text{ \AA}$, the Van der Waals' radius in graphite. (See Figs. 1 and 2.) The values of $r(\text{C})$ for rubidium and potassium lamellar compounds (3) are 1.682 \AA and 1.721 \AA , giving a small but systematic deviation for the series K, Rb, and Cs of +2.6%, +0.3%, and -4.5%.

The image-charge potential energy is given by

$$V_{\text{image}}(z) = - \frac{e^2}{4(z - z_o)} \quad (4)$$

At the equilibrium position, $V_{\text{image}} = -24.72 \text{ kK}$. The image-charge force is given by

$$\begin{aligned} F_z^{(\text{image})}(z) &= - \frac{dV_{\text{image}}(z)}{dz} \\ &= - \frac{e^2}{4(z - z_o)^2} \end{aligned} \quad (5)$$

which is just the coulomb force between two charges $+e$ and $-e$ separated by $2(z - z_o)$. If the surface has defects, that is, missing carbon atoms, the above approach must be modified (6). Let us suppose that there are M defects (circular holes in the conduction plane) located at (x_m, y_m) with radii a_m ($m = 1, 2, \dots, M$); then given the following functions,

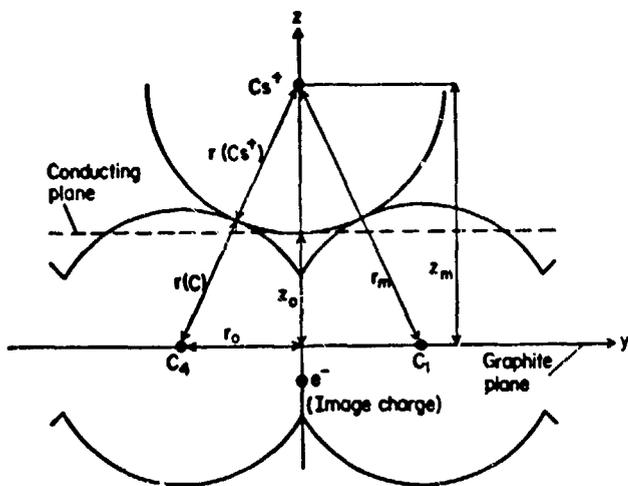


FIG. 1

Side view of bonding geometry of adsorbed cesium atom on graphite; C_1 and C_4 are carbon atoms as shown in Fig. 2, e^- is position of image charge (other symbols defined in text).

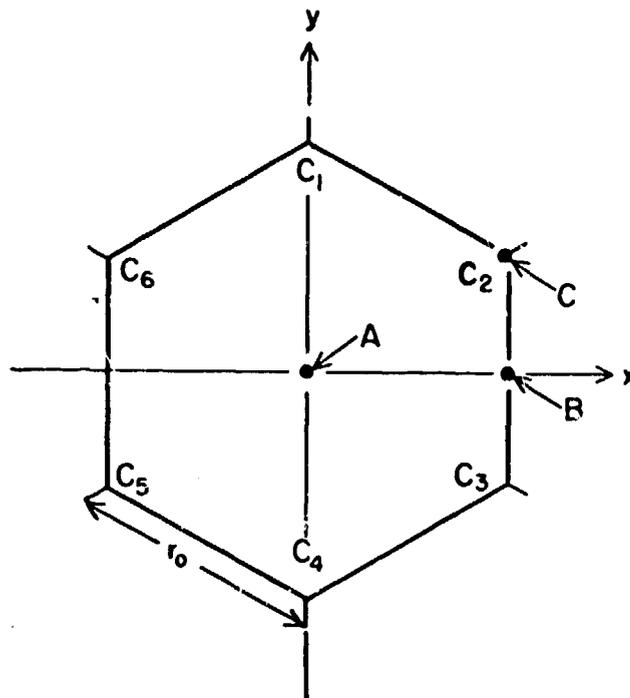


FIG. 2

Top view of bonding geometry of adsorbed cesium atom on graphite; site A is adsorption site (minimum of potential energy), site B is above C-C bond (saddle point in potential energy surface), and site C is above carbon atom (maximum of potential energy).

$$A(x,y) = \sum_{m=1}^M a_m^2 \exp \{ -[(x - x_m)^2 + (y - y_m)^2] / a_m^2 \} \quad , \quad (6)$$

and

$$A_\alpha(x,y) = \sum_{m=1}^M (x_\alpha - x_{\alpha m}) \exp \{ -[(x - x_m)^2 + (y - y_m)^2] / a_m^2 \} \quad , \quad (7)$$

where $\alpha = x$ or y , the potential energy of a charge $+e$ located at (x,y,z) in the presence of M defects in the conducting plane at $z = z_0$ can be approximated by:

$$V(x,y,z) = - \frac{e^2}{4[A(x,y) + (z - z_0)^2]^{1/2}} \quad , \quad (8)$$

and the forces are

$$F_{\alpha}(x,y,z) = \frac{e^2 A_{\alpha}(x,y)}{4[A(x,y) + (z - z_0)^2]^{3/2}} \quad (9)$$

for $\alpha = x$ or y and

$$F_z(x,y,z) = - \frac{e^2(z - z_0)}{4[A(x,y) + (z - z_0)^2]^{3/2}} \quad (10)$$

These expressions reduce to the image-charge expressions [eqs. (4) and (5)] far from defects, including the condition that F_x and F_y vanish. Further, from Eq. (9) it is clear that a defect is a scattering center from consideration of the bulk electrostatic interaction.

IV. Induction Interaction

The charge on the cesium ion can induce an instantaneous dipole moment in the carbon atoms of the graphite crystal (and vice versa), which leads to a potential energy contribution of the following form (7)

$$\phi^{(ind)}(R) = - \frac{C_4}{R^4} \quad (11)$$

where $\phi^{(ind)}$ is in units of $e^2/2a_0$ (e is the electronic charge and $a_0 = 0.5292 \text{ \AA}$ is the first Bohr radius), R is the interatomic separation in units of a_0 , and C_4 is given by

$$C_4 = q_1^2 \alpha_2 + q_2^2 \alpha_1 \quad (12)$$

where q_i is the charge (in units of e) on atom i and α_i is its polarizability (in units of a_0^3). The polarizability of Cs^+ can be estimated using Slater's screening constants (7). A better estimate is obtained by multiplying the observed value for iso-electronic xenon (8), $\alpha_{\text{Xe}} = 27.1 a_0^3$, by the ratio of α_{Cs^+} to α_{Xe} as calculated via the Slater method. The result is $\alpha_{\text{Cs}^+} = 17.4 a_0^3$. Since the polarizability of a molecule is the sum of its bond polarizabilities, and since carbon bond polarizability versus bond order is fairly linear, the polarizability of graphite per carbon atom can be obtained from the linear interpolation of the aliphatic (single bond, bond order $n = 1$) and aromatic (benzene, $n = 1-1/2$) bond polarizabilities. The polarizability of a graphite carbon atom is then $3/2 \alpha_{\text{C-C}}$ ($n = 1-1/3$), since the graphite bond order (5) is $1-1/3$ and there are $1-1/2$ such bonds per atom. The result is $\alpha_{\text{C}} = 10.8 a_0^3$. The electron donated by the cesium atom is shared by at least six nearest carbon atoms, so that the charge on any carbon atom is no more than $-e/6$. Therefore, taking $q_{\text{C}} = -1/6$ as being correct for the most important nearby carbon atoms, $C_4 = 11.26 \frac{e^2}{2a_0} \times a_0^4$. Define

$$A = \frac{C_4}{r_m^4} \quad (13)$$

the induction contribution per nearest carbon atom at the equilibrium position (position A in Fig. 2); then $A = 1.188$ kK.

V. Dispersion Interaction

Although the charge distribution in a free atom or ion is spherical, there are fluctuations due to the rapidly moving electrons that result in a net instantaneous dipole moment. This dipole can then induce an instantaneous dipole in another atom, leading to a potential energy contribution of the following form (7)

$$\phi^{(dis)}(R) = -\frac{C_6}{R^6} \quad (14)$$

where $\phi^{(dis)}$ is in units of $e^2/2a_0$, R in units of a_0 , and C_6 is given by the London-Pitzer formula (8)

$$C_6 = \frac{3}{2} \frac{\alpha_1 \alpha_1}{E_1^{-1} + E_2^{-1}}, \quad (15)$$

where α_1 is in units of a_0^3 and E_1 , the characteristic energy of atom 1, is in units of $e^2/2a_0$. Pitzer has shown empirically that E_1 is roughly twice the ionization potential. Since $E/E_1 = 2.27$ for neon and 2.39 for xenon (8), let us take $E_C = 2.27 E_I^{(C)} = 1.878 e^2/2a_0$ and $E_{Cs+} = 2.39 E_I^{(2)}(Cs) = 4.409 e^2/2a_0$. Therefore, $C_6 = 370.8 \frac{e^2}{2e_0} \times a_0^6$. Define

$$B = \frac{C_6}{r_m^6} \quad (16)$$

the dispersion contribution per nearest carbon atom at the equilibrium position; then $B = 1.010$ kK.

VI. Valence Interaction

At short range, the overlap of electron charge clouds of two atoms gives rise to a repulsive potential energy contribution of the form (7)

$$\phi^{(val)}(R) = b \exp(-aR/a_0), \quad (17)$$

where R is in units of a_0 and a is given by

$$a \left(\frac{e^2}{2e_0} \right)^{1/2} = E_I^{1/2}(1) + E_I^{1/2}(2), \quad (18)$$

with $E_I(i)$ in units of $e^2/2a_0$. $E_I^{(1)}(C) = 0.8273 e^2/2a_0$ and $E_I^{(2)}(Cs) = 1.845 e^2/2a_0$; hence, $a = 2.268$. Define

$$\alpha = a \frac{r_m}{a_0}, \quad (19)$$

then $\alpha = 14.11$ and

$$\phi^{(\text{val})}(R) = Q \exp \left[\alpha \left(1 - \frac{R}{r_m} \right) \right], \quad (20)$$

where Q is the valence contribution per nearest carbon atom at the equilibrium position. Q can be determined by requiring that the force on the cesium ion at position A, the adsorption site on a perfect graphite surface, be zero.

VII. Integral Corrections

Because the potential energy of interaction of the cesium ion with the free-surface graphite crystal converges slowly with distance into the crystal, an integral correction must be added to the discrete sum over graphite carbon atoms. In Fig. 3, the problem is indicated schematically as a spherical sector containing discrete atoms and a continuous distribution over the remainder of the crystal (free-surface bulk minus a crater).

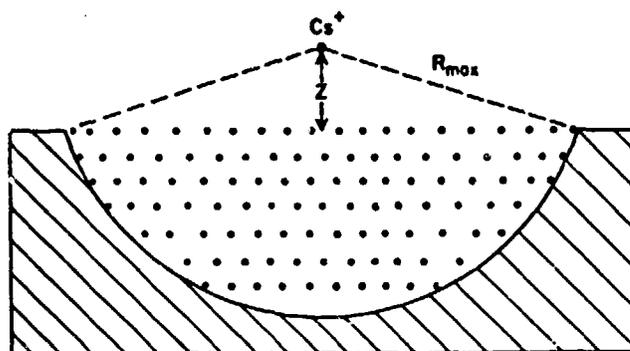


FIG. 3

Schematic representation of lattice summation and integral correction (continuous distribution) to potential energy of cesium ion over graphite surface.

The distance R_{max} is sufficiently large that only the induction and dispersion terms are appreciable. The integral correction for the potential term ϕ is

$$\delta\phi(R_{\text{max}}) = 2\pi\rho \int_{R_{\text{max}}}^{\infty} dR R(R - Z)\phi(R), \quad (21)$$

where ρ is the number density of graphite and Z is the height of the cesium ion over the graphite plane. Likewise, the z -gradient correction is

$$\delta\nabla_z \phi(R_{\text{max}}) = -2\pi\rho \int_{R_{\text{max}}}^{\infty} dR R\phi(R). \quad (22)$$

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VIII. Results

As outlined above, Q was calculated by summing over 8125 graphite carbon atoms within a radius of $R_{\max} = 33.1 \text{ \AA}$ and applying the integral corrections for the remainder of the graphite crystal. The resulting value of Q is 1.791 kK, which exceeds by about 0.2% the value obtained for an R_{\max} half as big (approximately one-ninth the number of carbon atoms). Q was also calculated, as a check, by summing over the six nearest carbons, integrating over a uniform surface distribution for the top graphite plane (minus a disk of six times the atomic surface area), and integrating over the bulk (minus the first graphite plane). The result was remarkably close to the "exact" lattice sum plus correction - about 3% too small (likewise for the magnitude of the adsorption energy). Table I shows results for the barriers to translational motion of a cesium ion on a perfect graphite surface. (The positions A, B, and C are shown in Fig. 2 and are defined

TABLE I

Barriers to Migration of Cesium on a Perfect Graphite Surface

Position, i ^a	z_i (\AA) ^b	$\frac{\phi_i}{k_B}$ (kK) ^c	$\frac{\phi_i}{k_B}$ (kK) ^d	$\frac{\phi_i - \phi_A}{k_B}$ (kK)
A	2.970	-61.65	-60.5	0
B	3.045	-59.95	--	1.70
C	3.055	-59.75	--	1.90

^aPositions A, B, and C shown in Fig. 2.

^bHeight of cesium ion above graphite surface for minimum in potential energy

^cPotential energy, $R_{\max} = 33.1 \text{ \AA}$ (k_B = Boltzmann's constant)

^dExperimental adsorption energy (9).

such that the total force on the cesium atom is zero). The kinetic energy at a temperature of about 1000 K or more is enough for a cesium ion to surmount the barriers on a perfect graphite surface; that is, the ion behaves essentially like a free particle. The experimental value for adsorption energy (9) was obtained from two adsorption isotherms of cesium on TS-688 graphite in the Henry's Law regime, i.e., where the Cs partial pressure is proportional to concentration. In view of the serious nature of the approximations herein employed, the close agreement with experiment should be viewed with some caution. Moreover, Zumwalt has pointed out that the true adsorption site is probably over a defect (10).

The effect of missing carbon atoms in the graphite surface ("holes" or "pits") upon

the adsorption energy of cesium is dramatic, as shown in Table II. The adsorption energy is given as a function of the size of the pit in the graphite surface. Three carbon-carbon bonds must be broken for the first missing carbon atom. (See Fig. 4.) The 1-atom

TABLE II

Adsorption Energy of Cesium over Holes in the Graphite Surface

Number of Missing C Atoms	Number of Broken C-C Bonds/Missing C Atom	$-\frac{\phi}{k_B}$ (kK) ^a	z (Å) ^b
0	0	61.3	2.97
1	3.00	60.8	2.92
2	2.50	63.2	2.61
4	2.25	70.4	2.11
6	2.00	102.0	1.40
24	1.75	99.0	1.26
∞	1.50	--	--

^aPotential energy, $R_{\max} = 14.5 \text{ \AA}$ ($k_B = \text{Boltzmann's constant}$).

^bHeight of cesium ion above graphite surface for minimum in potential energy.

hole is repulsive by one-half kilokelvin relative to the perfect surface. For the next carbon atom to be removed, two more bonds must be broken. (See Fig. 5.) The 2-atom hole is attractive by two kilokelvin. With four adjacent carbon atoms missing, three different shapes are possible (the one with trigonal symmetry is shown in Fig. 6). The 4-atom hole is attractive by about nine kilokelvin. A truly noticeable transition occurs when the hexagonally-symmetric 6-atom hole is made from the 5-atom hole by breaking only one more C-C bond.* (See Fig. 7.) At this point, the Cs^+ ion is just able to squeeze down comfortably into the hole at a height above the graphite surface not far from the conducting plane ($z_0 = 1.28 \text{ \AA}$). As the number of missing atoms is increased, very little effect is seen, though the minimum energy rises slightly by the 24-atom hole, the next hexagonally-symmetric hole after the 6-atom hole. (See Fig. 8.) The reason for the higher energy is that the Cs^+ can get close only to the carbon atoms at the edge of the 24-atom hole, while in the 6-atom hole, the Cs^+ is completely surrounded by carbon neighbors. The atoms below the first layer do not have a great effect on the potential energy,

* By this simplified thermochemical argument, we mean to illustrate the distinct energetic preference for 6- (or more) atom holes over a random distribution of smaller holes in a graphite surface (see Table II for number of C-C bonds broken per missing carbon atom as a function of the number of atoms comprising a hole).

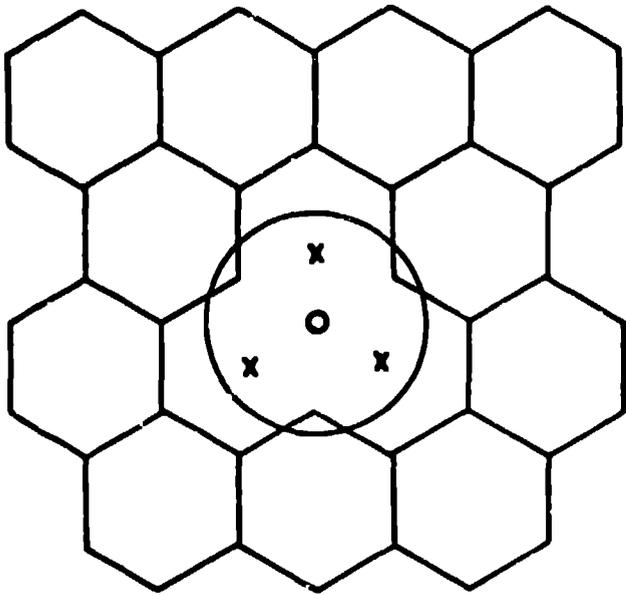


FIG. 4

1-atom defect in a graphite surface.
 (— = carbon-carbon bond with $r(C) = 1.421 \text{ \AA}$, o = position of missing carbon atom, x = equilibrium positions of Cs^+ ion, large circle with $r(\text{Cs}^+) = 1.69 \text{ \AA}$ shows size of ion compared with hole in graphite surface.)

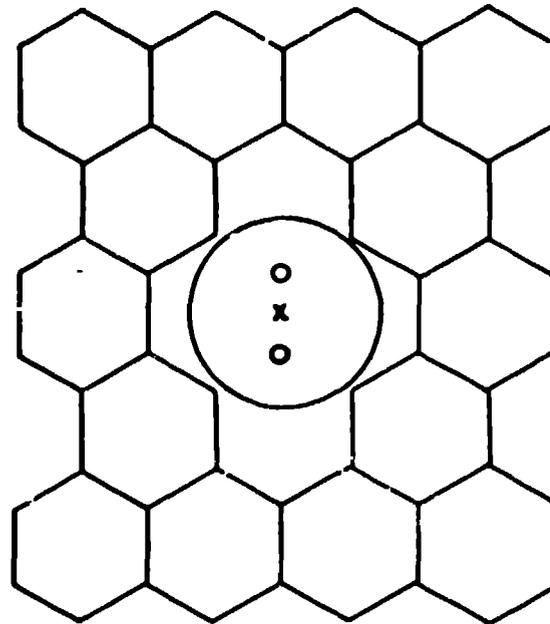


FIG. 5

2-atom defect in a graphite surface
 (same key as Fig. 4).

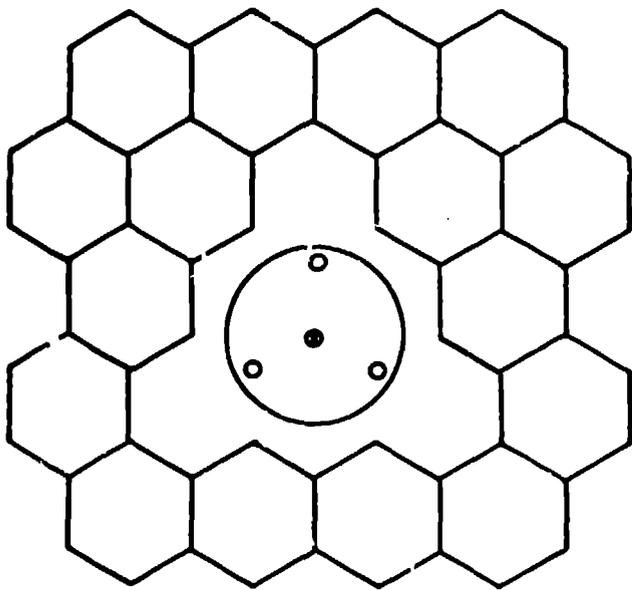


FIG. 6

4-atom defect in a graphite surface
 (same key as Fig. 4).

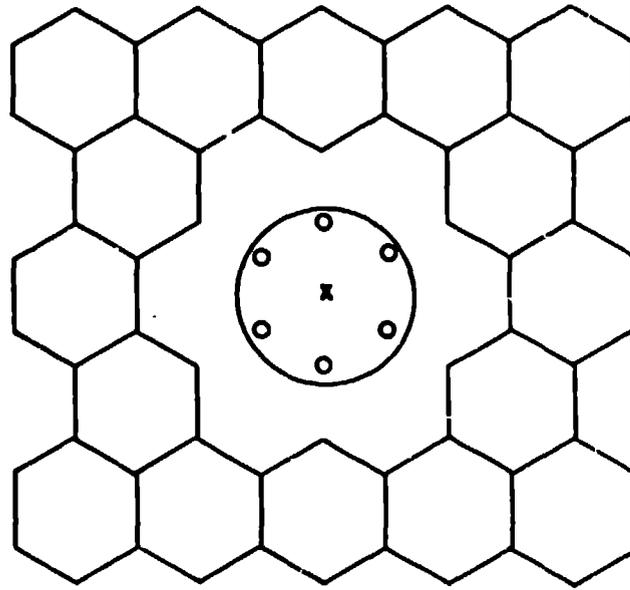


FIG. 7

6-atom defect in a graphite surface
 (same key as Fig. 4).

as verified by calculations of the 6-atom hole with one and four atoms missing from the sublayer. The conducting plane, which contains the donated Cs electron, serves to hold the Cs^+ at $Z \sim Z_0$ (see Eq. 10), at least in this model.

With regard to adsorption behavior itself, it should be noted that the nature of the interaction changes dramatically as the cesium atom leaves the surface and the ionization reaction of Eq. (3) is reversed to give a desorbed neutral cesium atom. This essentially quantum mechanical effect can be accounted for in a semi-empirical way by multiplying the appropriate ionic quantities (polarizability, charge, etc.) by an ionic character function $\chi(z)$ shown in Fig. 9, and the corresponding atomic neutral quantities by $1 - \chi(z)$.

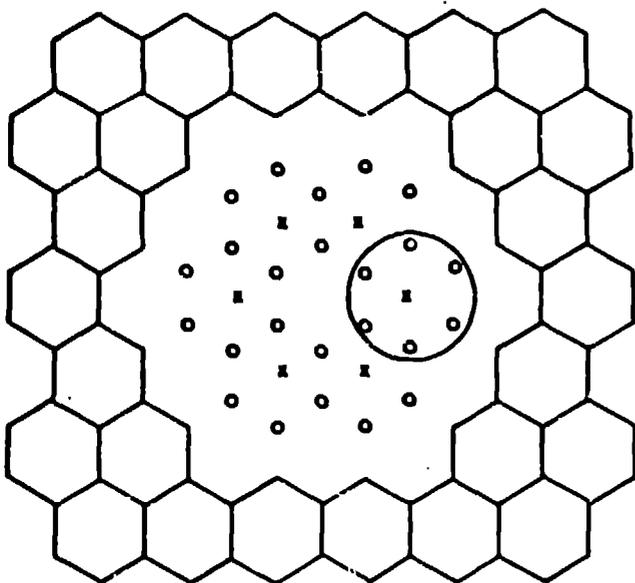


FIG. 8

24-atom defect in a graphite surface (same key as Fig. 4).

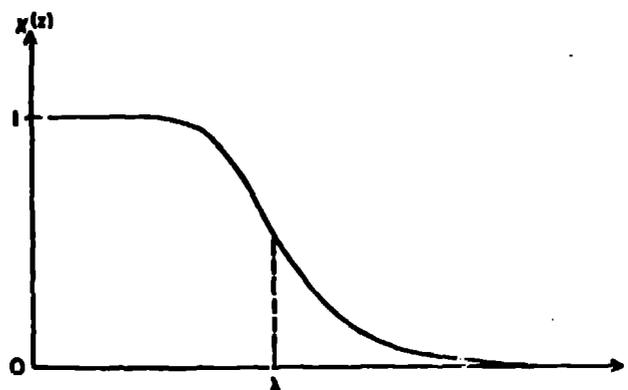


FIG. 9

Ionic character function $\chi(z)$ as a function of the distance z of the ion above the free surface of a crystal; λ is the characteristic length of χ .

Thus, a completely classical calculation, such as a molecular dynamics simulation could include implicitly this electronic rearrangement in the potential energy terms. The characteristic length λ of the ionic character function as well as its shape could, in principle, be inferred from quantum mechanical electronic calculations. The lithium-graphite system would be of some interest in this regard. Although the lithium atom is not spontaneously ionized by graphite, the substantial difference in the electronegativities of lithium and carbon guarantees that the lithium-graphite bond will have a great deal of ionic character, thus exhibiting much of the valence electronic distortion to be expected in the fully ionic cesium-graphite system.

Preliminary dynamical calculation results indicate that a cesium ion on a perfect graphite surface moves along the surface like a free particle at temperatures near 1000 K. Thus, truly diffusive or random-walk behavior at such temperatures requires the presence

of defects in the graphite surface. (Thermal motion of the carbon atoms in the graphite surface is probably not sufficient.) A molecular dynamics calculation of the thermal annealing of surface defects in graphite might show the coalescence of randomly distributed 1-, 2-, . . . , and 5-atom pits into 6- (or more) atom pits and thereby yield a realistic surface defect distribution (11). Then, with the methods outlined here, a surface diffusion coefficient could be calculated, essentially from first principles, and compared with the recent experimental results.

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