Antiproton storage materials: Response of neon, argon, and benzene to a negative particle

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The energies of systems composed of a point negative charge (e.g., antiproton, $\bar{p}$, or negative muon, μ^-) and an atom or small molecule (He,Ne,Ar,C_6H_6) have been calculated using large-basis-set, correlated *ab initio* electronic-structure methods. By invoking the Born-Oppenheimer approximation, these energies are used to generate potential surfaces for the interaction of a negative particle with an atom or small molecule. The results indicate that the particle-induced polarizations are significant even at 10-bohr separations in neon. Unusual behavior of the dipole moments of both argon and neon was calculated to occur as a function of separation. The molecular-orbital approach used here may also be useful in analyzing recent ionization behavior in low-energy $\bar{p}$–inert-gas scattering experiments. Furthermore, the calculated large degree of polarization further constrains the design of materials for long-term normal matter storage of antiprotons for advanced energy sources.

INTRODUCTION

Low-energy antiproton ($\bar{p}$)–inert-gas scattering experiments recently performed at the European Organization for Nuclear Research have resulted in the observation of unexpected ionization behavior.[1,2] These observations were examined theoretically,[2,3] and, in the case of helium, the theory was extended to much lower energies than were reported experimentally.[4] We undertook to examine the adiabatic[5] potential surfaces of the $\bar{p}$-Ne and $\bar{p}$-Ar systems via a molecular-orbital method in order to compare the response of these systems to that of the $\bar{p}$-He system which had been investigated using this method.[4] Furthermore, the calculations were extended to the considerably more complex case of the response of benzene (C_6H_6) to a nearby negative particle. The response of atoms and molecules to a nearby negative particle is of particular interest as a model for materials designed for normal matter storage of antiprotons. In this context the antiproton can be treated as a nonpolarizable point negative charge which is, for the most part, located outside the van der Waals radii of the atoms or molecules.

Several schemes have been suggested for the long-term storage and utilization of large amounts (milligrams to grams) of antimatter, particularly in relationship to advanced propulsion systems.[6] For example, small amounts (roughly 10^9 particles) of antiprotons can be confined by electric and/or magnetic fields in ion traps, but in the case of greater numbers of particles, the energy of the confining fields exceeds the total energy of the stored particles.[7] Storage of neutral antihydrogen (atoms and molecules) eliminates the Coulomb problem but presents challenges in the formation and control of neutral, polarizable atoms or molecules.[8] Storage of antiprotons within normal matter has been recently reinvestigated,[9] and appears to offer many opportunities.

Normal matter and antimatter are independently stable, but antimatter-normal matter interactions are essentially governed by Coulomb forces which lead to an-

nihilation. That the forces between matter and antimatter lead inescapably (in the absence of external fields) to annihilation can be demonstrated in several ways.[10] We have derived a particularly natural approach based on an extension of classical electrostatics to quantum-mechanical systems. Within the framework of classical electrostatics, we consider a movable test charge in a system of fixed nonpolarizable charges. The Poisson equation demands that the Laplacian of the potential energy of this system is zero everywhere, except within the fixed charges. It immediately follows that there are no maxima or minima outside the fixed charges. If, on the other hand, the test charge approaches a polarizable object, the resulting polarization lowers the potential energy at all radial separations and results in an attractive force contribution.[11] Since this force varies more rapidly with separation than r^{-2} (potential faster than $1/r$), the Laplacian becomes negative. As a simple example, the induced dipole moment μ of a linearly polarizable object is proportional to the electric field of the test charge. Thus μ varies as r^{-2}. But the field from a dipole varies as r^{-3}. It follows that the field due to the induced dipole at the location of the test charge is $E_d \propto \mu r^{-3} \propto r^{-5}$. The potential energy is proportional to $-r^{-4}$ (the negative sign indicates that the induced dipole always is in the direction which lowers the potential energy), for which the Laplacian is negative. Thus we conclude from this intuitive argument that potential-energy maxima may exist in free space, but to a first approximation, minima may only exist at the charges. This result can be extended to quantum-mechanical systems by applying the Hellmann-Feynman theorem to the Laplacian operator (see Ref. 12 and the Appendix). The conclusion applies directly to an antiproton near a lattice of fixed, but polarizable atoms: although local maxima in the potential energy as a function of the $\bar{p}$ coordinates may exist in the matter, local minima do not exist except on the nuclei. Furthermore, for such a negative antiparticle, quantum-mechanical barriers to annihilation with the matter nuclei do not ex-

ist. Since the required potential-energy minima do not exist, "equilibrium" or "static" storage is impossible. Recent calculational results suggest that "dynamic" or "active" storage on molecular dimensions may, however, be possible through the application of alternating electric fields.[13]

The purpose of this paper is twofold: (1) to calculate the effects of polarization as a negative particle approaches atoms and small molecules, and (2) to analyze how such polarization constrains the design of active storage media. The previous work had not included the effects of polarization on storage media, and from the polarization argument above, such storage schemes can be expected to be more difficult to design. The general approach was to apply molecular orbital methods to calculate the adiabatic potential of the antiproton plus atom-molecule system as a function of the separation of the two species. Calculations on the noble gases were performed with extended, uncontracted basis sets[14] using the Gaussian−86 system of programs.[15] (Semiempirical modified neglect of differential overlap[16] calculations failed even qualitatively to represent the polarization response). No attempt was made to calculate these systems when the particle-nucleus distance was less than a few tenths of a bohr; by this time a low-energy antiproton would be well past the point of no return and clearly uninteresting for particle storage. Calculations on the noble-gas atoms incorporated electron correlation at the MP4 (fourth-order Møller-Plesset[17]) level. Calculations of benzene were considerably more complex. The largest basis sets used were correlated calculations at the MP2/6-31g* level and SCF (self-consistent-field) calculations at the 6-311++g** level.[18] Electric potentials and fields were computed directly from the wave functions.

I. POTENTIAL SURFACES OF A NEGATIVE PARTICLE ($\bar{p},\mu^-$) PLUS NEON OR ARGON

Recent calculations on $\bar{p}$+He demonstrated that ionization occurs at a critical nucleus-particle separation in a manner reminiscent of critical dipole behavior.[5] The authors of the previous work suggested that this ionization process would be common to all atoms, but we thought it essential to investigate this phenomenon with heavier noble gases. Our calculations on He are consistent with the published $\bar{p}$+He work.

The energies of the $\bar{p}$+Ne/Ar systems are plotted in Fig. 1(a), as a function of internuclear separation and in Fig. 1(b) as a function of (separation)$^{-4}$. The nearly straight line behavior in Fig. 1(b) indicates that the atoms respond linearly to the electric field of a point charge, i.e., as a linearly polarizable system, down to distances approaching 2 bohr. The polarizabilities (α) of Ne and Ar can be calculated from this data if one assumes that the field at the atom due to the point charge is uniform. The calculated values for α in atomic units for Ne and Ar are 2.726 and 11.07; these compare very favorably with experimental values of 2.669 (Ref. 19) (2% error) and 11.074 (Ref. 20) (0.04% error). Nonlinear polarization is evident as the negative particle further approaches a Ne or Ar nucleus to a fraction of a bohr ($1/r^4$=10) as

shown in Fig. 1(c).

The energy of the highest occupied atomic orbitals (approximately the negative of the ionization potential) for Ne and Ar are shown in Fig. 2. Unlike the Ne, Ar is not ionized at any separation by a negative particle at rest; instead the energy of the highest energy orbital reaches a maximum −0.05 hartree near 1 bohr. The situation in Ne is more complex, with an electronic response that is virtually indistinguishable from ionization with these calculations. The peak orbital energy occurs at a particle separation of 0.8 bohr (−0.003 hartree). At smaller separations, calculations suggest that, like in Ar, all of the electrons of Ne+$\bar{p}$ are bound. This is consistent with the corresponding captured-particle limits of Ne+$\bar{p}$→F$^-$ and Ar+$\bar{p}$→Cl$^-$ in which the outer electrons of the ions are strongly bound.

The calculated dipole moments of Ne and Ar are plotted in Fig. 3 as a function of particle separation. The behavior of argon is very unusual in that the dipole has a maximum near 2 bohr. This is clearly not at the same particle separation as the point of minimum ionization potential which is closer to 1.25 bohr. This decrease in dipole moment does not, however, result in a decrease in the attractive force between the atom and the negative particle. An even more unusual variation in the dipole moment of neon has been calculated as a function of particle separation. The reversal that is calculated corresponds to the electron cloud of neon swinging around to the same side of the nucleus as the negative particle; however, again there is no decrease in the attractive force between the atom and the particle.

The net result of the particle-induced polarization is always strongly attractive. The unusual local maximum in the dipole moment which occurs without ionization is not likely to be limited to argon, and may exist in other atoms which are difficult to ionize, such as krypton and xenon.

The significance of these calculations to antiproton storage within matter lies in estimating the distances at which energies due to polarization of nearby atoms can overwhelm an externally applied confinement field. To achieve stability, the magnitude of the external field must be large with respect to the internal field arising from induced polarization, but less than the intrinsic dielectric strength of the materials (10^6–10^7 V/cm). Furthermore, given a storage cavity of molecular dimensions, the frequency of this field would need to be on the order of 10^{13} Hz, in the far infrared region of the electromagnetic spectrum. These values, in combination with the calculated polarization responses, suggest that minimum separations between atoms or molecules and antiprotons of 5 Å or more may be required to reduce induced fields to less than 10^6 V/cm for dynamic storage schemes even in systems as nonpolarizable as argon atoms.

II. POTENTIAL SURFACE OF A NEGATIVE PARTICLE ($\bar{p},\mu^-$) PLUS BENZENE

The electronic structure of the much more complex and interesting system $\bar{p}$ plus benzene was also studied. One reason benzene is interesting is because it has a large

zero-field quadrupole moment, which is such that nega-tive charges are repelled from the sixfold axis. Further-more, benzene is a fundamental building block of many organic molecules and yet its structure is simple enough to treat rigorously by electronic structure methods. Two factors contribute to the relatively large zero-field quad-rupole in this molecule: (1) the carbon atoms are slightly negative with respect to the hydrogen atoms and (2) the π system, which places electron density above and below the molecular plane. Our computed zero-field values for benzene's quadrupole moment are in agreement with pre-vious calculations[21] and experiment.[22] The potential en-ergy of a negative particle approaching the face of a ben-zene molecule, where the effects of polarization are neglected, is plotted in Fig. 4.

The total energy of a system composed of benzene and a negative particle approaching the molecule on axis is shown at different computational levels in Fig. 5. Calcu-lations at the MP2/6-311+ +g** level were not possible; therefore correlation effects must be estimated from the smaller 6-31g* basis set computations. These correlated calculations suggest that the correlation energy for $\bar{p}$+ benzene is greater than that of benzene itself and there-fore the barrier is probably less than the SCF barrier of 150 meV calculated at the 6-311+ +g** level and much less than the 270-meV barrier calculated using the less flexible 6-31g* basis set. These contracted basis sets are somewhat too small to fully represent the polarization effects due to a nearby charge, and therefore the true po-tential barriers are likely to be still lower and located far-
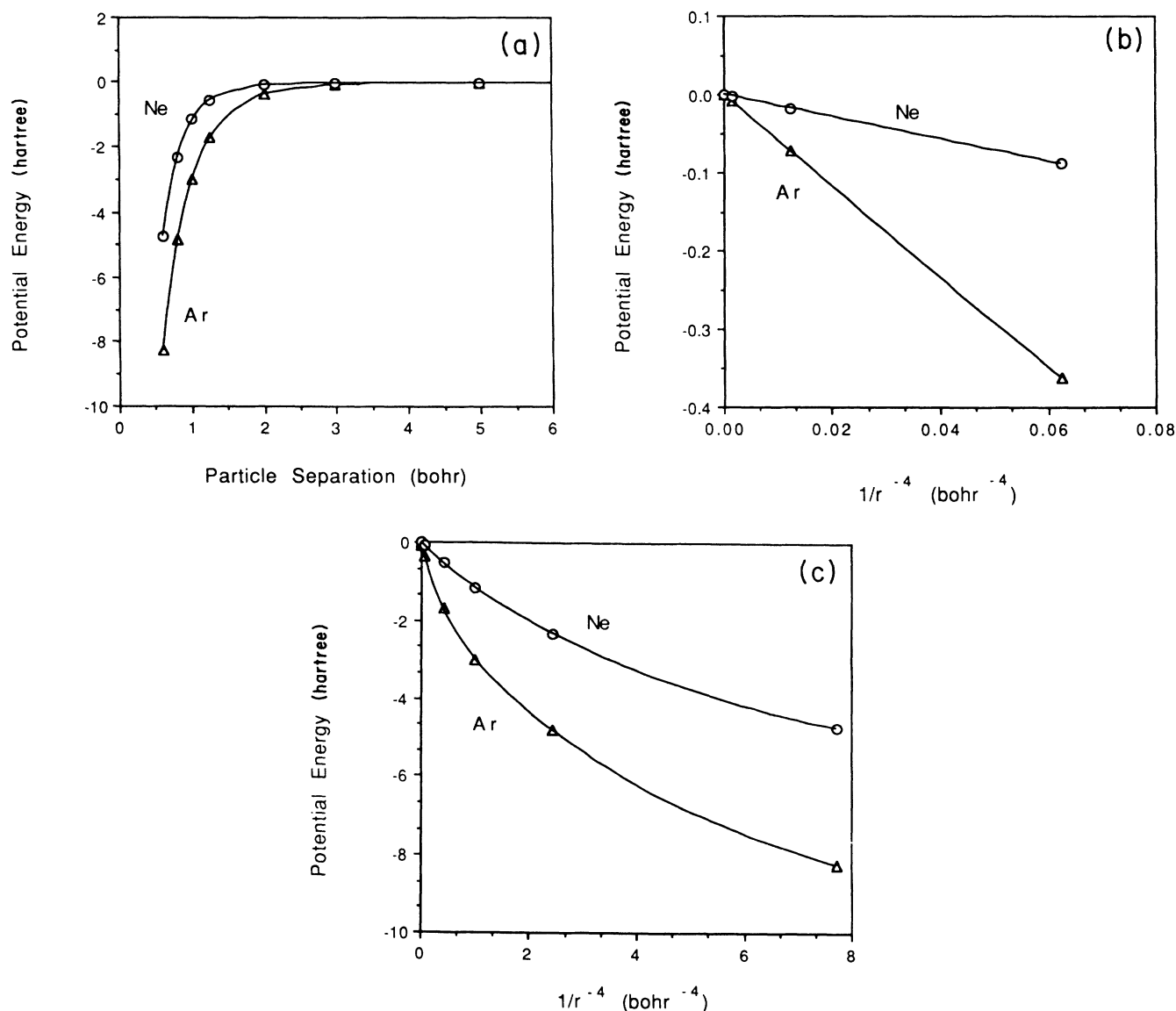

FIG. 1. (a) Potential energy of a negative particle near a Ne or Ar atom (relative to infinite separation) plotted linearly in distance. (b) Potential energy of a negative particle near a Ne or Ar atom (relative to infinite separation) plotted as (separation)$^{-4}$. The hor-izontal axis corresponds to distances from infinity to 2 bohr. (c) Same as (b), but the horizontal axis is extended to 0.6 bohr.
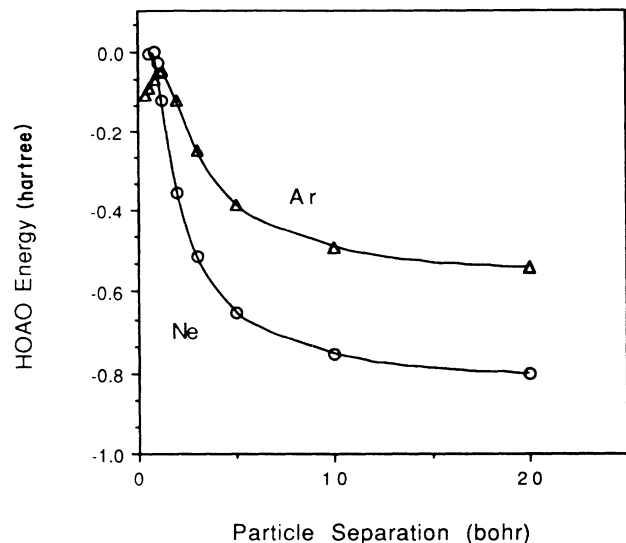
FIG. 2. Energy of the highest occupied atomic orbital of Ne or Ar as a function of distance from a negative particle. Positive values denote ionization.

FIG. 4. Potential energy of a negative particle above the ring plane of benzene and stacked benzene ("cyclophane") as a function of distance from the ring plane, *if polarization effects are not included* (calculated from zero-field electric potential).

ther from the ring than these calculations indicate.

Reoptimization of the C—C or C—H bond lengths at the SCF level in the presence of the charge showed less than 1% changes in bond lengths despite a calculated dipole moment of up to 3.3 D. The large induced dipole, which results as the π system is pushed away from the negative point charge, is what leads to a strong attractive force which overwhelms the zero-field quadrupole moment.

Calculations were also performed off axis. One calculation with the charge over a C—C bond and one with the charge over a C atom showed somewhat lower ener-

gies for the same plane-particle distance than for the high-symmetry (on-axis) case. This is consistent with the reduced off-axis zero-field quadrupole moment plotted in Fig. 4.

The barrier to approach of negative particles constrained to the symmetry axis is not sufficiently high and deep to prevent tunneling of all but the lowest energy (meV) particles as massive as an antiproton or antideuteron. Calculations on stacked rings, as in compounds such as the cyclophanes, indicate that the zero-field quadrupole is substantially larger (Fig. 4). These calculations incorporated two planar benzene rings stacked face to
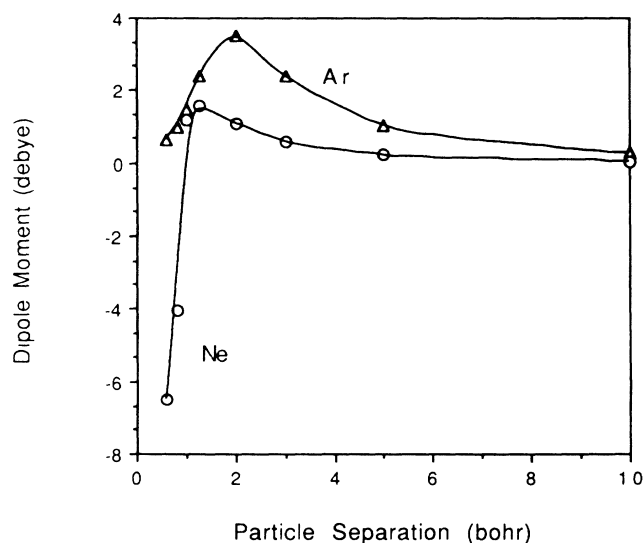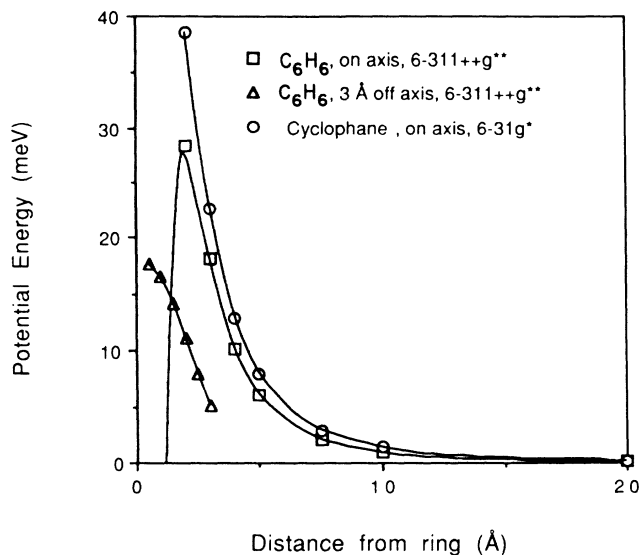

FIG. 3. Dipole moment of Ne or Ar as a function of distance from a negative particle. Markedly different behavior is calculated for the two atoms.
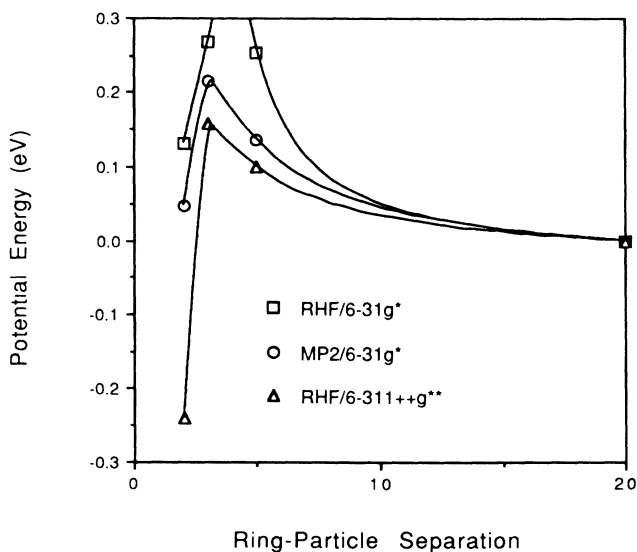

FIG. 5. Potential energy of an axial negative particle near benzene as a function of distance from the ring plane. Curves for three computational levels are plotted.

face at a ring plane separation of 3.2 Å, a crude approximation to the cyclophanes which have slightly bent benzene rings. These systems were too large to compute except with the 6-31g* basis set, but do show an enhancement of the zero-field quadrupole moments. This may imply a higher barrier to negative particle approach along the sixfold axis.

Finally, the zero-field quadrupole moments of several trisubstituted rings were calculated in order to estimate the effect of substituents on the local fields. The quadrupole moments in these cases contained contributions from the charge distributions within the substituents which are difficult to separate from the quadrupole due to the π system alone. Enhancement of the quadrupolar potential by 50% was seen in tri-(SiH$_3$) substituted benzene, however. This is 30% larger than that calculated for the stacked benzene rings.

III. CONSEQUENCES FOR STORAGE OF ANTIPROTONS

The large polarization effects calculated above emphasize the severe constraints placed on the design of normal matter materials for dynamic storage. As pointed out in Sec. I, achieving confinement when nearby atoms are as nonpolarizable as argon is already difficult, and storage becomes progressively more difficult in media of higher polarizability. Polarization effects can be minimized in two ways: by utilizing structures of low inherent polarizability and by maximizing the void volume of the storage medium, thereby increasing the average separation between matter and antimatter. Low inherent polarizability can probably be best obtained by using low-Z elements and by avoiding ionic structures (which have relatively highly polarizable anions).

Maximization of the void volume is a more difficult task. Crystal lattice defects are not large enough, given the calculated overwhelming polarization effects. Zeolites may deserve study because of the large void dimensions (≈ 10 Å). These compounds are composed of strongly polar bonds mainly between silicon, aluminum, and oxygen, and are therefore likely to be highly polarizable. Smaller voids are probably available through the synthesis of spheroidal organic molecules, perhaps similar to buckminsterfullerene C_{60}.[23] C_{60} itself is probably too polarizable because its structure is fully aromatic and therefore similar to (conductive) graphite. Other molecules of similar or larger dimension (C_{60} is roughly 8 Å in diameter) and lower polarizability should be synthesizable.

Storage molecules composed of molecules with large quadrupole moments similar to benzene, if linked so that each segment is electronically isolated, may facilitate storage by flattening the potential surface in certain directions within the void volume. Our calculations on benzene indicate that an axial particle-induced dipole overwhelms the zero-field quadrupole moments in benzene at distances of 2 Å. The barrier is only 0.10–0.15 eV; however, the minimum energy for a particle constrained to the sixfold axis of benzene is calculated to be -9.14 eV at the center of the ring.

CONCLUSIONS

Large, uncontracted basis-set correlated molecular-orbital calculations on Ne and Ar indicate significant differences in the response of these atoms to the approach of a negative particle ($\bar{p}$ or μ^-). Neon's response is virtually indistinguishable from ionization at this level of calculation and its dipole moment reverses sign at a critical separation; Ar clearly does not ionize but shows a maximum in its dipole moment at a critical separation. These calculations may be of use in interpreting antiproton-inert gas scattering experiments.

Similar calculations on benzene with the particle constrained to benzene's sixfold symmetry axis are much more complex. Benzene's zero-field quadrupole is overwhelmed by the particle induced dipole at a distance of approximately 2 Å from the ring plane. An axial barrier of only 0.1–0.15 eV or less is presented by the molecule. however.

These calculations serve to place additional constraints on the design of materials for dynamic antiproton storage. The exceptionally large polarizations induced by a nearby negative particle have not previously been considered in such schemes. The conclusions are that polarization effects must be included in any dynamic storage scheme and furthermore that cavity dimensions of many angstroms will be required to avoid overwhelming polarization effects that lead to annihilation. Atomic size voids or lattice defects are not likely to be sufficiently large for particle storage.

Two nonporous types of materials have been suggested. One class of material comprises the zeolites (clays), which are principal aluminosilicate cages. The alternate class of materials comprises molecules as yet unsynthesized, but which are related to compounds now studied as simple guest-host complexes. These molecules may have smaller voids than the zeolites, but may be less polarizable or may assist in confining a particle through potentials from zero-field dipole or higher moments.

APPENDIX

We outline the proof of instability of a heavy negative particle interacting with normal ground-state matter. The Born-Oppenheimer (BO) Hamiltonian, obtained by neglecting the kinetic energies of the nuclei and antiproton, is

$$H = H^{BO} = T^e + V^N + V^{N\text{-}e} + V^{e\text{-}e} + V^{N\text{-}\bar{p}} + V^{e\text{-}\bar{p}} . \qquad (A1)$$

$v^{N\text{-}\bar{p}}$ and $V^{e\text{-}\bar{p}}$ are the Coulombic interactions of the nuclei and electrons with the antiproton. T^e, V^N, $V^{N\text{-}e}$, and $V^{e\text{-}e}$ are the electron kinetic energy and Coulombic nuclei-nuclei, nuclei-electron, and electron-electron interactions.

The BO electronic states are assumed known,

$$H\psi_n = W_n \psi_n \ , \tag{A2}$$

$$\langle \psi_m | \psi_n \rangle = \delta_{mn} \ , \tag{A3}$$

$$\psi_n = \psi_n(\{\mathbf{r}_i\} | \{\mathbf{R}_i\}, \mathbf{x}) \ , \tag{A4}$$

$$W_n = W_n(\{\mathbf{R}_i\}, \mathbf{x}) \ , \tag{A5}$$

where the parametric dependence of the electronic wave functions and eigenvalues on all the heavy particle coordinates $\{\mathbf{R}_i\}$ and $\mathbf{x}$ is indicated in Eqs. (A4) and (A5). The BO eigenvalue W_n is the potential energy surface on which the nuclei and antiproton move.

The stability of any configuration of the antiproton and nuclei can be deduced by examination of the Laplacian operator with respect to antiproton coordinate $\mathbf{x}$ as it acts on the ground state BO surface W_0. A negative Laplacian indicates an unstable position.[10,24] The eigenvalue is given by

$$W_0 = \langle \psi_0 | H | \psi_0 \rangle \ , \tag{A6}$$

where the coordinates are suppressed, with the angle brackets denoting integration over all electron space. Using the Hellman-Feynman theorem, or equivalently by applying ∇_x to Eqs. (A6) and (A3) and combining, we have

$$\nabla_x W_0 = \langle \psi_0 | \nabla_x H | \psi_0 \rangle \ . \tag{A7}$$

The application of $\nabla_x \cdot$ to Eq. (A7) gives

$$\nabla_x^2 W_0 = \langle \psi_0 | \nabla_x^2 H | \psi_0 \rangle + \langle \nabla_x \psi_0 \cdot | \nabla_x H | \psi_0 \rangle$$
$$+ \langle \psi_0 | \nabla_x H \cdot | \nabla_x \psi_0 \rangle \ . \tag{A8}$$

There are two types of terms to consider; the first term on the right-hand side of Eq. (A8) is easy to evaluate. The argument is as follows: $\nabla_x^2 H$ contains only two of the potential terms in Eq. (A1), namely,

$$\nabla_x^2 H = \nabla_x^2 (V^{N\text{-}\bar{p}} + V^{e\text{-}\bar{p}})$$

$$= (e^2/4\pi\epsilon_0)\nabla_x^2 \left[-\sum_{i=1}^N \frac{Z_i}{|\mathbf{R}_i - \mathbf{x}|} + \sum_{i=1}^n \frac{1}{|\mathbf{r}_i - \mathbf{x}|} \right]$$

$$= -(e^2/\epsilon_0) \left[-\sum_{i=1}^N Z_i \delta^3(\mathbf{x} - \mathbf{R}_i) \right.$$

$$\left. + \sum_{i=1}^n \delta^3(\mathbf{x} - \mathbf{r}_i) \right] \ . \tag{A9}$$

Thus the first term in Eq. (A8) evaluates to be

$$\langle \psi_0 | \nabla_x^2 H | \psi_0 \rangle = -e^2 \epsilon_0^{-1} \rho^e(\mathbf{x} | \{\mathbf{R}_i\}, \mathbf{x}) \ , \tag{A10}$$

as long as $\mathbf{x} \neq \mathbf{R}_i$ (antiproton not on top of a nucleus). ρ^e is the non-negative electron density in the matter, evaluated at the position of the antiproton. This term contributes to a negative Laplacian.

The other two terms in Eq. (A8), of the form $\langle \psi_0 | \nabla_x H \cdot | \nabla_x \psi_0 \rangle$, must be evaluated by expansion in the full BO eigensystem, Eqs. (A2)–(A5). First, one applies ∇_x to the BO Schrödinger equation (A2) and projects with the basis. This gives

$$\langle \psi_m | \nabla_x \psi_n \rangle = \langle \psi_m | \nabla_x H | \psi_n \rangle / (W_n - W_m) \ , \tag{A11}$$

valid for $W_m \neq W_n$. Equation (A11) is used to construct an expansion for $\nabla_x \psi_0$. The ground-state electronic wave function may be assumed real and nondegenerate, which eliminates the problems with $n = m$ in Eq. (A11). This expansion for $\nabla_x \psi_0$ is substituted into the second and third terms on the right-hand side of Eq. (A8) to give

$$\langle \nabla_x \psi_0 \cdot | \nabla_x H | \psi_0 \rangle + \langle \psi_0 | \nabla_x H \cdot | \nabla_x \psi_0 \rangle$$
$$= 2 \sum_{m \neq 0} \mathbf{Z}_{om} \cdot \mathbf{Z}_{om}^* / (W_0 - W_m) \ , \tag{A12}$$

$$\mathbf{Z}_{om} = \langle \psi_0 | \nabla_x H | \psi_m \rangle \ . \tag{A13}$$

This term is negative as long as $W_0 - W_m < 0$ or $W_m > W_0$, which is true for the ground electric state. Thus we have proven that $\nabla_x^2 W_0$ is nonpositive for any configuration of the heavy particles in the BO approximation for the ground electronic state of matter.

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[1]L. H. Andersen, P. Hvelplund, H. Knudsen, S. P. Møller, K. Elsenser, K.-G. Rensfelt, and E. Uggerhøj, Phys. Rev. Lett. **57**, 2147 (1986).

[2]L. H. Anderson, P. Hvelplund, H. Knudsen, S. P. Møller, A. H. Sørensen, E. Elsenser, K.-G. Rensfelt, and E. Uggerhøj, Phys. Rev. A **36**, 3612 (1987).

[3]P. D. Fainsteil, V. H. Ponce, and R. D. Rivarola, J. Phys. B **21**, 2989 (1988).

[4]M. Kimura and M. Inokuti, Phys. Rev. A **38**, 3801 (1988).

[5]W. J. Hehre, L. Radom, P. V. R. Schleyer, and J. A. Pople, *Ab Initio Molecular Orbital Theory* (Wiley, New York, 1986), p. 12.

[6]G. Vulpetti, J. Br. Interplanet. Soc. **39**, 391 (1986).

[7]C. F. Driscoll, in *Low Energy Antimatter*, edited by D. B. Cline

(World Scientific, Singapore, 1986), p. 184.

[8]R. L. Forward, in Ref. 7, p. 47.

[9]L. J. Campbell, in *Proceedings of the Antiproton Science and Technology Workshop, Singapore, 1987*, edited by B. W. Augenstein (Rand Corporation, 1988), p. 342.

[10]S. Earnshaw, Trans. Cambridge Philos. Soc. **7**, 97 (1842).

[11]This is equivalent to stating that the response of the polarizable object is in the direction which lowers the potential energy.

[12]The derivation of this result is presented in the Appendix. In summary, application of the Hellmann-Feynman theorem to the Laplacian operator using the Born-Oppenheimer states of the system leads to three terms. One term represents the electron density at the position of the negative particle and this

term is always less than zero. The other terms involve gradients of the wave function and of the Hamiltonian. These terms can be shown to be less than zero *for the electronic ground state of the system.* It is not possible, however, to derive the classical $1/r^4$ potential for an induced dipole from these terms. Since all three terms are negative for a ground electronic state, quantum-mechanical barriers to annihilation of an antiparticle do not exist for molecules in their ground state.

[13]D. H. Dunlap and V. M. Kenkre, Phys. Rev. B **34**, 3625 (1986).

[14]An uncontracted ($13s9p5d$) basis set was used for neon; see G. Maroulis and D. M. Bishop, Chem. Phys. Lett. **114**, 182 (1985). For argon, an uncontracted ($16s10p5d$) basis set was used; see G. Maroulis and D. M. Bishop, J. Phys. B At. Mol. Phys. **18**, 4675 (1985).

[15]Frisch *et al.*, *Gaussian–86* (Carnegie-Mellon Quantum Chemistry Publishing Unit, Carnegie-Mellon University, Pittsburgh, 1986).

[16]M. J. S. Dewar and W. Thiel, J. Am. Chem. Soc. **99**, 4899 (1977).

[17]C. Møller and M. S. Plesset, Phys. Rev. **46**, 618 (1934).

[18]R. Poirier, R. Kari, and I. G. Csizmadia, *Handbook of Gaussian Basis Sets* (Elsevier, New York, 1985).

[19]R. P. Saxon, J. Chem. Phys. **59**, 1539 (1973).

[20]R. R. Teachout and R. T. Pack, At. Data **3**, 195 (1971).

[21]Our SCF 6-311++g^{**} calculations give $Q = -9.19$ D Å, compared to the full CI value of -8.78 D Å; see T-K. Ha, Chem. Phys. Lett. **79**, 313 (1981).

[22]The experimental value is -8.69 D Å; see M. R. Battaglia, A. D. Buckingham, and J. H. Williams, Chem. Phys. Lett. **78**, 421 (1981).

[23]H. Kroto, Science **242**, 1139 (1988).

[24]J. H. Jeans, *The Mathematical Theory of Electricity and Magnetism* (Cambridge University Press, New York, 1908), p. 165.