Calculation of the ground-state energy V_0 of quasifree positrons in rare-gas fluids

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The energy V_0 of the bottom of the conduction band (relative to vacuum) of quasifree positrons in rare-gas fluids is calculated as a function of fluid density. The calculations are performed within the framework of the Wigner-Seitz approximation [Phys. Rev. **43**, 804 (1933)] for nonpolar fluids, using a semiempirical analytical potential to model the positron–rare-gas-atom interactions. For all the rare gases studied, V_0 is negative and decreases almost linearly with increasing density. Extended to the solid-phase density range, our V_0 calculations are in good agreement with available experimental data for rare-gas crystals.

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INTRODUCTION

The recent developments in the use of positrons as a quantitative probe of matter in all its forms stimulate the interest of theoretical investigations concerning the positron states in gases, liquids, and solids [1]. The fundamental problem to be solved concerns the form of the interaction of the positron with matter, which is different from that for the electron. This is primarily associated with the difference in charge, but there is also a difference due to the fact that there is no Fermi sea of positrons in the target and therefore the exchange part of the interaction potential is not present. For this reason, positron-matter studies provide unique insight into the fundamental aspects of interactions of charged particles with matter both theoretically and experimentally. During its finite lifetime (typically, $\sim 10^{-10}$ s), the positron can be observed in delocalized, trapped, or positronium (Ps) states, depending on its energy and on the nature of its interaction with the medium. For positrons injected into simple fluids or solids with a wide range of low incident energies (below the Ps-formation threshold), only elastic scattering is dominant. Because of this reason, the rare-gas fluids and solids traditionally serve as initial test cases for novel theoretical and experimental methods involving scattering of either electrons or positrons. The two quantities of central interest for characterizing quasifree electron or positron states in nonpolar fluids are the ground-state energy V_0 (relative to vacuum) and the effective mass m^* of injected particles. A great deal of effort has continually been devoted to the description of electronic transport properties in those fluids [2]. In previous papers, we have calculated the dependence of V_0 and m^* of excess electrons in rare-gas fluids as a function of fluid density n [3–8]. In this work, we calculate $V_0(n)$ in the case of quasifree positrons in fluid helium, neon, argon, krypton, and xenon, using the Wigner-Seitz (WS) model [9] for nonpolar fluids.

POSITRON-ATOM INTERACTION

To model properly the positron-fluid interaction, the choice of a good positron-atom potential is of utmost importance. In the present paper, we describe the positron-atom interaction by means of a semiempirical analytical potential based on the analytical Dirac-Hartree-Fock-Slater atomic screening function $\phi(r)$ tabulated by Salvat *et al.* [10] in the following parametrized form:

$$\phi(r)=\sum_{i=1}^{3} A_i \exp(-b_i r) , \tag{1}$$

where r is the distance of the positron from the atom. The parameters A_i and b_i are given in the paper of Salvat *et al.* [10] for all the atoms considered here. The function $\phi(r)$ describes the screening of the nuclear charge Ze by the atomic electrons. The positron-atom potential can thus be expressed as

$$V_{e^+-at}(r)=\frac{Ze^2\phi(r)}{4\pi\epsilon_0 r}+V_{corr}(r) , \tag{2}$$

where e is the positron charge and ϵ_0 is the permittivity of vacuum. The first term represents the Coulomb potential produced by the bare atomic nucleus screened by the atomic electrons in their unperturbed ground-state configuration. The correlation term $V_{corr}(r)$ takes into account the effects of target polarization induced by the positron. In contrast to electron-atom systems, no exchange interaction is present.

It is well known that in positron-atom collisions correlation effects are very difficult to describe adequately without involving any adjustable parameter. Asymptotically, as for the case of electronic systems, the correlation potential reduces, at large distances, to the simple polarization form

$$V_{corr}(r)=-\frac{e^2\alpha}{2(4\pi\epsilon_0)^2 r^4} , \tag{3}$$

TABLE I. Values of the atomic polarizability α and of the adjustable parameter ρ (see text) used in the calculations. SL denotes the positron scattering length obtained using the positron-atom potentials with these parameter values. σ is the atomic hard-sphere diameter used in the determination of the atom-atom pair-correlation function $g_a(r)$ (see text). α is given both in atomic units (a.u.) and in SI units (C m^2/V). a_0 is the Bohr radius.

Quantity	Helium	Neon	Argon	Krypton	Xenon
α (a.u.)	1.383[a]	2.670[a]	11.07[a]	16.77[a]	27.29[a]
α (10^{-41} C m^2/V)	2.309	4.456	18.49	27.99	45.56
ρ (units of a_0)	0.1272	0.1413	0.1794	0.1978	0.2176
SL (units of a_0)	-0.470	-0.558	-4.11	-7.75	-26.7
σ (units of a_0)	4.830[b]	5.195[b]	6.435[b]	6.803[b]	7.748[b]

[a] Reference [16].
[b] Reference [17].

where α is the atomic polarizability. The difficulty arises when the positron is near the target atom. A simple way to circumvent this difficulty is to introduce in Eq. (3) a cutoff function $w(r)$ depending on some adjustable parameter. The semiempirical correlation potential proposed by Nakanishi and Schrader [11] allows one to describe well the positron-atom interactions in the gas phase. This correlation potential has the following form:

$$V_{\rm corr}(r)=-\frac{e^2\alpha}{2(4\pi\epsilon_0)^2r^4}w\left[\frac{r}{\rho}\right], \tag{4}$$

where

$$w(x)=[1-e_8^x\exp(-x)]^2, \tag{5}$$

ρ is an adjustable parameter, and e_8^x represents the power series for the exponential function, truncated after the eighth power of x [11].

In this paper, we adopt this type of correlation potential. The parameter ρ is adjusted to reproduce the experimentally determined positron scattering length. In cases where the experimental scattering length is not available, the choice of this parameter is made from comparisons of our calculated cross sections with experiment [12–15]. The values retained for ρ are listed in Table I for the different rare-gas atoms studied, along with those of α and of the calculated scattering length.

POSITRON-FLUID INTERACTION

In the Wigner-Seitz (WS) model [9], each atom in the fluid is replaced by an equivalent atomic sphere of radius $r_{\rm WS}$, defined by

$$\frac{4\pi}{3}r_{\rm WS}^3=\frac{1}{n}. \tag{6}$$

In the sphere, the short-range positron-atom interactions are described by the atomic potential $V_{e^+-{\rm at}}(r)$. The effect of the fluid density occurs mainly through the screened long-range positron-atom polarization interactions. Since the positions of the atoms are correlated, these interactions are related to the atom-atom pair-correlation function $g_a(r)$ of the fluid. The potential is assumed spherically symmetric, which, when combined with the average translational symmetry condition, amounts to neglecting fluctuations in the fluid and considering an ensemble-average potential acting on the positron [9,18].

At a point $\mathbf{r}$ inside the WS sphere around an atom located at $\mathbf{r}=0$, the total potential $V_{e^+-l}(r)$ seen by the positron can be expressed as the sum of the potential produced by the atom at the origin and of the ensemble-averaged potential produced by the atoms lying outside the sphere [4,6,8]

$$V_{e^+-l}(r)=V_{e^+-{\rm at}}(r)+n\int V_{e^+-{\rm at}}(|\mathbf{r}-\mathbf{r}'|)F(|\mathbf{r}-\mathbf{r}'|)g_a(r')d\mathbf{r}', \tag{7}$$

where $F(r)$ is a screening function of the fluid that accounts for the effect of the field of the induced dipoles of the surrounding atoms. It is approximated by [18]

$$F(r)=\begin{cases}1, & r\le r_{\rm WS}\\ \left[1+\dfrac{2n\alpha}{3\epsilon_0}\right]^{-1}, & r>r_{\rm WS}.\end{cases} \tag{8}$$

The integration in Eq. (7) is over all the space excluding the WS sphere with the additional condition that the positron is assigned to only one sphere at once, that is, $|\mathbf{r}-\mathbf{r}'|>r_{\rm WS}$. The pair correlation function $g_a(r)$ is obtained by solving the Percus-Yevick equation [19] for a hard-sphere fluid model [20] with an atomic hard-sphere diameter given in Table I.

The ground-state energy V_0 of the quasifree positron is determined by solving numerically the Schrödinger equation with the interaction potential $V_{e^+-l}(r)$ of Eq. (7), and by subjecting the ground-state positron wave function $\Psi_0(r)$ to the WS periodic boundary condition $[d\Psi_0(r)/dr]=0$ at $r=r_{\rm WS}$.

RESULTS AND DISCUSSION

The results of our calculations of the ground-state energy V_0 of quasifree positrons in all studied rare gases are shown in Figs. 1 and 2 as a function of fluid density n. In Fig. 1, we compare the values of $V_0(n)$ obtained for helium and neon. As one can see, they show a very similar variation with the density; $V_0(n)$ is negative in both cases and decreases almost linearly with increasing n. This can

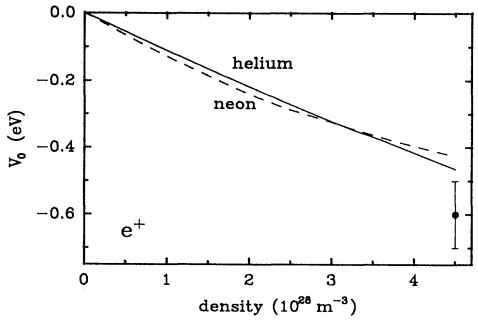

FIG. 1. Conduction-band energy minimum V_0 as a function of fluid density n for quasifree positrons in helium (solid line) and neon (dashed line). ●, experimental result obtained by Gullikson, Mills, and McRae (Ref. [21]) for crystalline neon at 4 K. For helium, the density at the critical point (5.19 K) is 1.05×10^{28} m^{-3} (Ref. [22]). For neon, the densities at the critical (44.4 K) and triple (24.55 K) points are 1.44×10^{28} m^{-3} and 3.72×10^{28} m^{-3}, respectively (Ref. [22]).

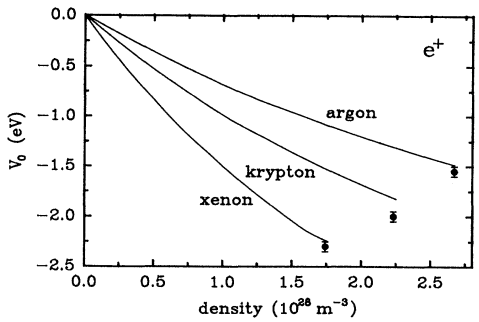

FIG. 2. Conduction-band energy minimum V_0 as a function of fluid density n for quasifree positrons in argon, krypton, and xenon. ●, experimental results obtained by Gullikson, Mills, and McRae (Ref. [21]) in crystalline samples at 4 K. The densities at the critical and triple points are, respectively, 0.80×10^{28} m^{-3} (150.7 K) and 2.13×10^{28} m^{-3} (83.8 K) for argon, 0.66×10^{28} m^{-3} (209.5 K) and 1.75×10^{28} m^{-3} (115.8 K) for krypton, and 0.51×10^{28} m^{-3} (289.7 K) and 1.36×10^{28} m^{-3} (161.4 K) for xenon (Ref. [22]).

readily be understood since the positron scattering lengths associated with the two atomic potentials are negative and nearly equal, and since the polarizabilities of helium and neon are also quite similar. We can thus expect comparable negative slopes of $V_0(n)$ near the origin [23] and similar screenings of the long-range polarization interactions. On the same figure, the experimental estimate of Gullikson, Mills, and McRae [21] in crystalline neon, deduced from positron Bragg diffraction measurements, is also shown for comparison. When extended to densities corresponding to the solid phase, our calculated values of V_0 for neon are slightly above the experimental estimate. It is worth noting here that, for the case of electrons, positive values of V_0 are observed for both helium and neon [3,4].

Figure 2 shows a comparison of our $V_0(n)$ results for argon [24], krypton, and xenon, along with the experimental estimates of Gullikson, Mills, and McRae [21] in the crystals. As one can see, there is a very good agreement between our solid-phase $V_0(n)$ calculations and these latter data. In contrast to what is observed for the density dependence of the ground-state energy of excess electrons in heavy rare-gas fluids [2,5–7], we do not see any minimum of $V_0(n)$ for the case of positrons; the position of the bottom of the positron conduction band changes only monotonically as a function of n. As for helium and neon, $V_0(n)$ is negative in all cases and for all the densities considered. Unfortunately, there are no experimental data of V_0 in the fluid phase with which to compare our results.

Recently, Puska and Nieminen [25] calculated the positron band structures, and also the positron work functions ($-V_0$), in rare-gas solids. They proposed a new semiempirical positron-atom correlation potential depending only on the polarizability of the atom and on a universal cutoff parameter r_1, valid for all rare gases and chosen by fitting the calculated positron band gaps to the measured ones. The positron work functions calculated by these authors [25] were found to be larger than the experimental ones, with the largest discrepancy, 1.4 eV, in the case of neon. Puska and Nieminen [25] suggested that their indirect determination of V_0 for positrons could be at the origin of this discrepancy. However, it seems that this problem should more probably be connected with the use of an incorrect value of the parameter r_1. In fact, the value of r_1 chosen by these authors does not reproduce the experimental positron-atom scattering length and total elastic scattering cross sections at very low energies [12–15]. This is particularly important since the work function is very sensitive to the value of the scattering length. The experimental increasing trend in the positron work function when going from neon to xenon can also be well explained by the systematic increase of the scattering length. We hope that this paper will contribute to motivate experimentalists to pursue experimental investigations on the conduction-band energies of quasifree positrons in rare-gas fluids.

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