Muon—alpha-particle sticking probability in muon-catalyzed fusion

D. Ceperley and B. J. Alder
Lawrence Livermore National Laboratory, University of California, Livermore, California 94550
(Received 19 November 1984)

The Green's-function Monte Carlo method is used to calculate the ground-state wave function of the muonic-molecular ion composed of a negative muon bound to a deuteron and a triton. Using the sudden approximation, the probability that the muon will remain bound to the escaping alpha particle after fusion occurs is found to be 0.90%, about 25% smaller than previous estimates based on the Born-Oppenheimer approximation. The numerical method for determining the wave function is discussed in detail.

I. INTRODUCTION

There has been considerable interest in catalyzing fusion in deuterium-tritium mixtures with negative muons because recent experiments have found that a single muon can cause about one hundred fusion reactions.[1] If the only relevant rates were those governing the formation of the muonic ion and the decay of the muon, approximately one thousand fusions would be catalyzed. However, for about 1% of the fusions, the muon sticks to the outgoing alpha particle and is prevented from catalyzing further fusions. This sticking mechanism limits the efficiency of the process. Recent measurements[2] of the sticking probability are about half of the theoretical estimates based on the Born-Oppenheimer approximation.[3,4] In this paper the sticking probability is calculated without this approximation.

The sticking probability ω is defined as the probability that in the reaction

$$(dt\mu)^+ \rightarrow (\mu^4\mathrm{He})^+ + n + 17.59\ \mathrm{MeV}\ ,$$

the muon will remain bound to the alpha particle. We restrict the initial state of the muonic molecule to the ground state. Because fusion occurs on a much faster time scale than muonic-molecular motions, the sudden approximation can be used to determine this probability, once the initial Φ_i and final Φ_f muonic wave functions are known at the instant of fusion, namely, at the coalescence point (taken to be the origin of the coordinate system) where the deuteron and triton are on top of each other. Under the sudden approximation the sticking probability is

$$\omega = \left| \int d^3r\, \Phi_i(r)\Phi_f(r) \right|^2 . \tag{1}$$

The final state is, of course, simply a neutron and a moving hydrogenlike ion. However, the initial state is a genuine three-body system. Let $\Theta(r_\mu, r_d, r_t)$ be the full three-body wave function in the coordinate system where the center of mass is at rest. Then the initial muonic wave function is

$$\Phi_i(r_\mu) = \frac{\Theta_i(r_\mu,0,0)}{\left[\int d^3r\, |\Theta_i(r,0,0)|^2 \right]^{1/2}} . \tag{2}$$

It has been previously assumed that one can make the Born-Oppenheimer (BO) approximation[3,4] for Θ_i and then Φ_i reduces to the ground-state wave function for a hydrogenlike ion with a helium nucleus of mass 5. However, because the mass of the muon is not small compared to the masses of the nucleons, this is not, as we shall see, a very good approximation. Variational calculations of the Hylleraas type could[5] be used to calculate the muonic wave function. However, a variational function is usually accurate only in the region where the probability density is high, not at the coalescence point. One expects those calculations to converge exceedingly slowly there with increasing number of terms in the expansion of the wave function. Monte Carlo calculations do not have this limitation.

Monte Carlo calculations have been used to calculate ground-state properties for a wide variety of quantum systems.[6] They are typically most useful when the number of degrees of freedom is too large for other methods to handle. However, the method is quite applicable to few-body problems as well. Although the Monte Carlo scheme to calculate wave-function values has been known for some years, the emphasis has always been on obtaining accurate energies. In the next section we will discuss the application of the Green's-function Monte Carlo (GFMC) method to this problem and introduce an improved method for calculating wave-function values. Section III will summarize the results for the muon—alpha-particle sticking probability.

II. NUMERICAL METHODS

The particular form of the GFMC method applicable to Coulombic systems has been discussed in detail in a recent paper.[7] We will assume the reader is familiar with that paper and follow its notation. The units of length used here will be muonic radii ($a_\mu = 0.256\times10^{-10}$ cm). The energies will be given either in muonic hartrees (5.6265 keV) or, when discussing binding energies [i.e., energies relative to the unbound system $(t\mu)+d$], in electron volts. The Hamiltonian in muonic atomic units for this system is

$$H = -D_\mu\nabla_\mu^2 - D_t\nabla_t^2 - D_d\nabla_d^2 - 1/r_{\mu d} - 1/r_{\mu t} + 1/r_{dt}\ , \tag{3}$$

where the "diffusion constants" in these units are $D_\mu=0.5$, $D_d=0.028\,17$, and $D_t=0.018\,81$.

A. The importance function

Essential to an accurate solution with the Monte Carlo method is an accurate importance function. Although the calculated expectation value of any property is independent of this function, the statistical variance is proportional to its error. Since we are calculating the value of the wave function at the coalescence point, the importance function must be reasonable in that region. Since this muonic ion is very similar to a hydrogen molecule, we use an importance function of the pair-product form that was successfully employed[8] for H_2:

$$\Psi_T(R)=g_{dt}(r_{dt})g_{d\mu}(r_{d\mu})g_{\mu t}(r_{\mu t})\tilde{g}(r_{dt}) \times[g_0(r_{\mu d})+g_0(r_{\mu t})] , \tag{4}$$

where $g(a,r)=\exp[-a_1/(a_2+r)-a_3 r]$ and $\tilde{g}(a,r)=\exp[-a_1/(a_2+r^2)]$. The analytic values for the derivative of the wave function at the point where two particles coalesce can be used to eliminate some of the free parameters and insure that the importance function has the proper behavior there, giving

$$\frac{1}{g_{ij}}\frac{dg_{ij}(r)}{dr}\bigg|_0=\frac{a_1}{a_2^2}-a_3=\pm\frac{0.5}{D_i+D_j} . \tag{5}$$

These cusp conditions are applied to the first three factors in Eq. (4) only and the cusp value for both g_0 and $\tilde{g}$ is assumed to be zero. The importance function contains a sum over the two orbital functions g_0, describing the alternating binding of the muon to the two nucleons. In principle, the parameters in these two orbitals could be different, but for convenience we have kept them the same. The additional function $\tilde{g}$ has been added to improve the importance function in the coalescence region.

After taking into account the cusp conditions, there are ten free parameters in the importance function. The values of these parameters are determined by minimizing the variational energy using configurations derived from a variational Monte Carlo[6] calculation. A very strong check on the correctness of the computer code is that the final answers be independent of the importance function within the statistical errors.[9] Accordingly we have used two sets of variational parameters (denoted I and II, their values are given in Table I). The variational energy for both of these trial functions is the same, namely, -0.5361 ± 0.0002. This corresponds to a binding energy of 305 ± 1 eV which is about 14 eV above the exact ground-state energy.

B. Green's-function Monte Carlo

The first step in calculating wave-function values is to calculate the ground-state energy. Although this has been done previously to the needed accuracy by both variational[5] and nonvariational[10] methods, it is instructive to reproduce those results by GFMC. The GFMC algorithm[6] finds ground-state properties by applying the operator

$$G(R,R')=\Psi_T(R)\langle R\,|\,[1+\tau(H-E_T)]^{-1}\,|\,R'\rangle\Psi_T^{-1}(R') \tag{6}$$

many times to the initial distribution $f_1=|\Psi_T(R)|^2$ leading to

$$f_{n+1}(R)=\int dR'\,G(R,R')f_n(R') , \tag{7}$$

when R refers to the nine-dimensional vector of particle positions, τ is the generational time step,[7] and E_T is the trial ground-state energy.[6] The convolution required in this equation is performed stochastically by using branched random walks. Thus the distribution f_n, referred to as the nth generation, consists of an ensemble of configurations $\{R\}$ and the convolution is interpreted as sampling, conditional on those configurations, a new ensemble of configurations from the operator (or matrix in configuration space) $G(R,R')$. This can be done exactly and an efficient method has been developed for Coulombic systems.[7]

The ground-state energy calculated with GFMC is $-0.538\,61\pm0.000\,04$ which corresponds to a binding energy of 319.2 ± 0.2 eV. This compares favorably with a recent variational calculation[5] of the Hylleraas type of 319.06 eV and a nonvariational calculation[10] of 319.15 eV.

C. Calculation of relative wave-function values

Repeated application of the evolution operator G can also give wave-function values.[11] Consider the total popu-

TABLE I. Trial wave-function parameters, variational energies, normalization, and overlap integrals for the two importance functions used. All lengths are in muon radii, energies in muon hartrees.

	Function I			Function II		
	a_1	a_2	a_3	a_1	a_2	a_3
g_{dt}	13.37	1.063	1.194	17,90	1.220	1.377
$g_{\mu t}$	−1.15	1.202	0.166	−1.14	1.199	0.169
$g_{\mu d}$	−1.28	1.256	0.135	−1.25	1.240	0.136
g_0	0.83	1.016	0.813	0.83	1.011	0.814
$\tilde{g}_{dt}$	−0.0575	0.230		−0.359	0.188	
E_v	−0.5360±0.0002			−0.5362±0.0002		
C_T	0.394±0.002			0.402±0.002		
N_T	864.1			17 124.0		
O_T	0.9980±0.0007			0.9989±0.0006		

lation after n applications of the Green's function to a single point R_0 in the first generation, i.e., define

$$P_n(R_0)=\int dR\, f_n(R)\ , \tag{8}$$

where $f_1(R)=\delta(R-R_0)$. Then using the eigenfunction expansion for G, it is easy to show that asymptotically

$$\lim_{n\to\infty} P_n(R_0)=\frac{\Theta(R_0)}{\Psi_T(R_0)}\frac{\int dR\,\Psi_T(R)\Theta(R)}{[1+\tau(E_0-E_T)]^{n-1}}\ , \tag{9}$$

where Θ and E_0 are the ground-state eigenfunction and eigenvalue, respectively. Unless the trial energy E_T is chosen exactly equal to the ground-state energy, the population will tend to grow or decline asymptotically depending on whether E_T is larger or smaller than E_0. The uncertainty in E_T does not significantly affect the calculated wave-function values because the energy is always calculated more accurately than wave-function values. Relative wave-function values are calculated by starting off a number of systems at any desired configuration (R_0) and finding the average number that asymptotically result. In practice the walks are followed for a sufficient number of generations until their distribution matches that of the ground state. To get absolute wave-function values, the normalization constant must be determined as discussed in Sec. II D.

Statistical fluctuations can be reduced by an order of magnitude using the following transformation: apply the inverse operator to G {i.e., $\Psi_T[1+\tau(H-E_T)]/\Psi_T$} to both sides of Eq. (7) and integrate over R. Then using Green's identity for H and defining the local energy as $E_L(R)=\Psi_T^{-1}H\Psi_T-E_T$, we obtain

$$P_n(R_0)=P_{n+1}(R_0)+\tau\int dR\, E_L(R)f_{n+1}(R)\ . \tag{10}$$

Using this relation recursively we obtain

$$P_n(R_0)=\prod_{i=2}^{n}(1+\tau\langle E_L\rangle_i)^{-1}\ , \tag{11}$$

where $\langle E_L\rangle_i$ is the average of the local energy over all walks which reach the ith generation.

In GFMC, fluctuations in branching arise for two reasons, namely, because one cannot sample G perfectly and because the importance function does not equal the ground-state wave function. In practice it is found[7] that the fluctuations in branching and hence in the total population are an order-of-magnitude larger than those due to the importance function alone. The above local energy estimate of the wave function, however, is only sensitive to importance-function errors, leading to a much lower variance. Figure 1 illustrates the convergence of the population and a comparison of the two estimators for walks starting at the triple coalescence point [i.e., $P_n(r_d=r_t=r_{\mu t})$]. Within the error bars[9] both estimators give the same results but the local-energy-estimator error bars are five times smaller.

Table II contains the results for various values of the muon distance from the coalescence point. From 10^4 to 10^5 walks have been started from each initial point and a generational time step of $\tau=0.5$ was used. The wave-

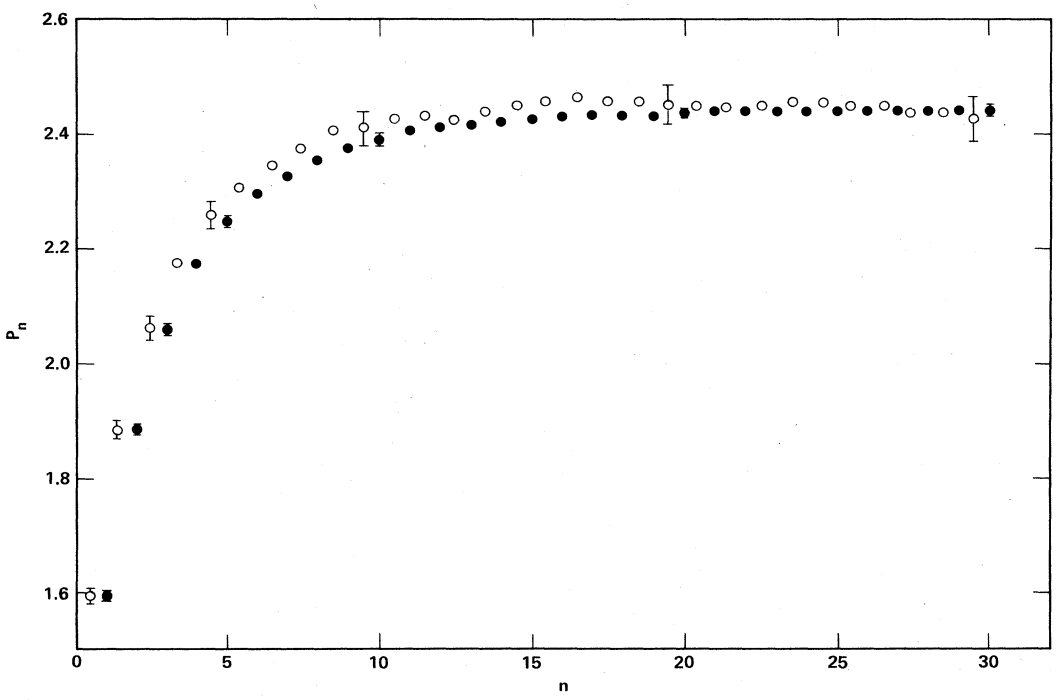

FIG. 1. The growth of the population vs the number of Monte Carlo generations using the population estimator (○) and the local energy estimator (●) at the triplet coalescence point (all three particles starting at the origin). The ○'s are shifted one-half generation to the left for clarity.

TABLE II. Normalized wave-function values $\Theta(r)$ at the coalescence point as a function of the muon distance r computed using two different importance functions. The numbers in parentheses are exponents. $P_{50}(r)$ is the population after 50 generations (computed with the local energy estimator) and re is the relative error of P_{50} and Θ.

	Function I			Function II		
r	$P_{50}(r)$	re	$\Theta(r)$	$P_{50}(r)$	re	$\Theta(r)$
0.0	2.447	0.005	1.264(−3)	0.852	0.003	1.265(−3)
0.5	2.313	0.004	5.072(−4)			
1.0	1.967	0.005	2.066(−4)			
1.5	1.676	0.004	8.963(−5)			
2.0	1.385	0.004	3.904(−5)			
2.5	1.130	0.004	1.715(−5)			
3.0	0.922	0.007	7.46(−6)			
3.5	0.758	0.004	3.46(−6)			
4.0	0.628	0.010	1.59(−6)	0.220	0.010	1.61(−6)
4.5	0.516	0.004	7.32(−7)			
5.0	0.425	0.010	3.38(−7)			
6.0	0.298	0.012	7.51(−8)			
7.0	0.211	0.015	1.70(−8)			
8.0	0.148	0.013	3.83(−9)	0.0527	0.017	3.87(−9)

function values are accurate to 0.5% for $r_\mu < 4$ and 1.0% for larger r. The error bars basically reflect the amount of computer time expended at each point. Importance function II has somewhat larger error bars for the same length of run but their results agree rather well.

There are three sources of systematic errors in this calculation, all of which are small. We have already mentioned that the ground-state energy must be known accurately. Note that the statistical error of 4×10^{-5} in the trial energy will give an error of 0.1% in the wave-function values after 50 generations, somewhat less than their statistical errors. Secondly, the local energy estimator for the wave function will contain some bias because the mean value of a product is not equal to the product of the mean values. (The original population estimator does not have such a bias.) This bias is, however, very small because of the large number of walks contributing to each term. We can estimate this bias by dividing up the sample various ways and arrive at an estimate to the relative bias of one-tenth of the statistical error. Finally, convergence of the wave-function values versus generation number must be obtained. The walks are followed for 50 generations since by then it is found that the average deuteron-triton distance reaches its asymptotic value and the wave-function values have stabilized.

The above method of calculating wave-function values has the desirable "zero-variance" property.[6] By that is meant that in the limit where the importance function approaches an eigenfunction of H, the variance of the calculated wave-function values goes to zero. But independent of the importance function, the GFMC method always yields the exact result within its statistical error bars provided the run is long enough that convergence has been obtained. There is no particular difficulty associated with computing wave-function values at the coalescence point. The local energy is initially on the order of unity, but it quickly decays to zero once the two nuclei have drifted apart.

D. Normalization

Calculation of absolute wave-function values requires two additional Monte Carlo calculations. Normalization is not necessary to find the muon—alpha-particle sticking probability since the initial muon-state functions are renormalized anyway [see Eq. (2)]. However, normalized wave-function values are necessary in order to compare results from different importance functions, to compute other types of matrix elements (such as the fusion rate), and to compare with other variational calculations.

Initially the importance function is normalized. For some simpler importance functions this step is unnecessary. Define the constant C_T by

$$\frac{1}{C_T}=\frac{1}{\Omega N_T}\int_\Omega dR\ |\Psi_T(R)|^2\ , \tag{12}$$

where we assume the molecule is confined in a (very large) box of volume Ω because the center of mass is not otherwise fixed. Variational Metropolis Monte Carlo[6] is used to compute the average:

$$C_T=\frac{\int dR\ \Psi_T^2(\Psi_A/\Psi_T)^2}{\int dR\ \Psi_T^2}=\langle(\Psi_A/\Psi_T)^2\rangle\ , \tag{13}$$

where Ψ_A is the function

$$\Psi_A=g_{dt}(r_{dt})\tilde{g}(r_{dt})e^{-\alpha[r_\mu-(r_d+r_t)/2]^2} \tag{14}$$

and N_T is its normalization

$$N_T=\frac{1}{\Omega}\int dR\ \Psi_A^2=4\pi(\pi/2\alpha)^{3/2}\int_0^\infty r^2dr\ g_{dt}^2(r)\tilde{g}^2(r)\ . \tag{15}$$

Care must be taken to prove that the variance of the estimator of Eq. (13) exists. In the present case this is equivalent to showing that the function Ψ_A^2/Ψ_T is bound-

ed for all values of its arguments. The constants C_T and N_T for the two trial functions are given in Table I.

The second step in calculating absolute wave-function values is to compute the overlap integral O_T [i.e., the integral in the numerator of Eq. (9)]. The calculation proceeds exactly as in Sec. II C but the initial condition is changed, namely, the first generation consists of points sampled from the square of the importance function. Then, as in Eq. (9), the population asymptotically becomes

$$O_T=\frac{\left[\int \Psi_T\Theta\right]^2}{\int \Psi_T^2}=\lim_{n\to\infty} P_n(\Psi_T^2)\ . \tag{16}$$

Note that the overlap with the ground state (see Table I) is almost unity, showing that our importance function overall is accurate. However, as we shall see in the next section, this does not imply individual importance-function values are accurate. Using the results of the calculation of O_T and C_T we can now normalize the wave-function values. The relative error of the normalization is only 0.27% so the normalized wave-function values do not have appreciably larger errors than relative wave-function values.

III. RESULTS AND CONCLUSIONS

Figure 2 shows the exact wave function divided by the Born-Oppenheimer value. Table II lists numerically the wave-function values at the coalescence point as a function of the muon distance. Since the adiabatic approximation assumes that the muon responds instantaneously to nuclear motions, the Born-Oppenheimer function is not spread out enough.

The wave-function values have been used to compute

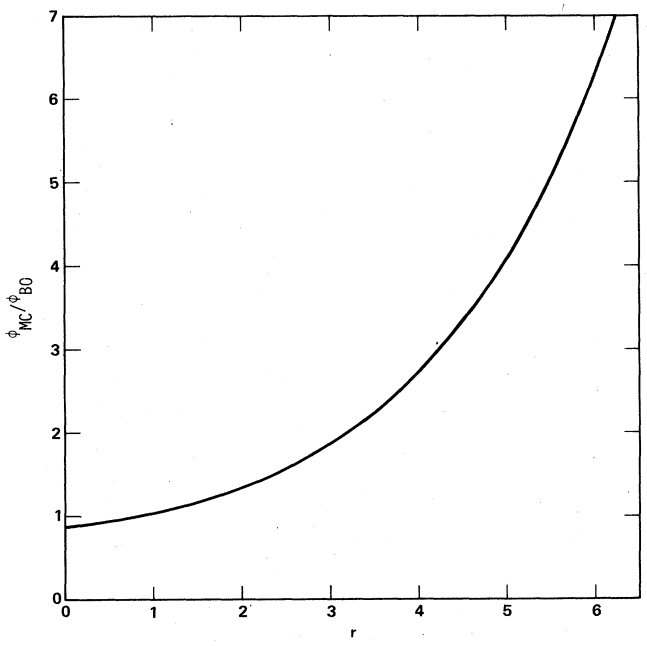

FIG. 2. The ratio of the exact wave function to the BO wave function (i.e., an exponential) both normalized as in Eq. (2) at various muon distances from the fused nuclei.

TABLE III. Comparison of the Monte Carlo determined values of the muon–alpha-particle sticking probabilities (given in percent) to various final states and the sum over all states, with those from the Born-Oppenheimer approximation. "Other" is the sum over all states not listed.

State	Monte Carlo	BO
$1s$	0.689	0.9024
$2s$	0.099	0.1288
$2p$	0.024	0.0321
$3s$	0.030	0.0391
$3p$	0.009	0.0116
$4s$	0.013	0.0166
Other	0.031	0.0405
Total	0.895±0.004	1.1711

the sticking probabilities in the various final states. The final-state wave function is

$$\Theta_f(r_\mu,r_\alpha,r_n)=e^{ip(R_c-r_n)}Y_{lm}(\hat{r}_{\alpha\mu})R_{ln}(r_{\alpha\mu})\ , \tag{17}$$

where p is the outgoing neutron momentum, $R_{ln}Y_{lm}$ is the Coulomb wave function, and R_c is the center of mass of the alpha-particle–muon atom. Setting the nuclear coordinates to zero as given in Eq. (2), averaging over the direction of p, and summing over m values, we obtain the sticking probabilities in the various final states:

$$\omega_{ln}=(2l+1)4\pi\left|\int_0^\infty r^2dr\,\Phi_i(r)j_l(qr)R_{ln}(r)\right|^2\ , \tag{18}$$

where $qa_\mu=5.844$.[12] They are given in Table III. Our values are consistently 24% lower than those from the BO function. This brings them into much better agreement with experiment.[2] Exact comparison to experiment is difficult as the measurements are at a finite density and collisions with the surrounding molecules strip off some of the muons [estimated to be (24±4)%].[3,4] The principal contribution to the integrand comes from values of r less than one, so the relative sticking probability is approximately equal to the square of the function plotted in Fig. 2 evaluated at the origin.

This calculation can be extended straightforwardly to the other initial states of the mesic molecule in a different symmetry class from the ground state, i.e., to the ($J=1$, $\nu=0$) and ($J=2$, $\nu=0$) states, by using the methods already developed for fermion systems.[13] However the fusion cross section in these states is small because the centrifugal barrier inhibits fusion[14] and so they are not important to the muon-catalyzed fusion process. The extension to the more interesting states ($J=0$, $\nu=1$) and ($J=1$, $\nu=1$) is more difficult because these states must be explicitly orthogonalized to the ground state otherwise any admixture of the ground state in the importance function will eventually dominate the numerical simulation.

ACKNOWLEDGMENTS

We wish to thank David Morgan for suggesting this problem. This work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

[1]S. Jones *et al.*, Phys. Rev. Lett. **51**, 1757 (1983).
[2]S. Jones, Proceedings of the Ninth International Conference on Atomic Physics, 1984 (unpublished).
[3]S. S. Gershtein *et al.*, Zh. Eksp. Teor. Fiz. **80**, 1690 (1981) [Sov. Phys.—JETP **53**, 872 (1981)].
[4]L. Bracci and G. Fiorentini, Nucl. Phys. A **364**, 383 (1981).
[5]A. K. Bhatia and R. J. Drachman, Phys. Rev. A **30**, 2138 (1984).
[6]D. M. Ceperley and M. H. Kalos, *Monte Carlo Methods in Statistical Physics,* edited by K. Binder (Springer, New York, 1979), p. 145.
[7]D. Ceperley, J. Comput. Phys. **51**, 404 (1983).
[8]D. Ceperley and B. J. Alder (unpublished).
[9]Statistical errors are computed by dividing the total computer run into about twenty blocks. Errors quoted are always our estimate of one standard deviation.
[10]S. I. Vinitskii *et al.*, Zh. Eksp. Teor. Fiz. **79**, 698 (1980) [Sov. Phys.—JETP **52**, 353 (1981)].
[11]K. S. Liu, M. H. Kalos, and G. V. Chester, Phys. Rev A **10**, 303 (1974).
[12]This muonic momentum is slightly different from those found in Refs. 3 and 4. We have used in 1977 atomic mass values with the relativistic kinematic formula to determine the outgoing momentum.
[13]D. M. Ceperley and B. J. Alder, Phys. Rev. Lett. **45**, 566 (1980).
[14]L. N. Bogdanova, V. E. Markushin, V. S. Melezhik, L. I. Ponomarev, Yad. Fiz. **34**, 1191 (1981) [Sov. J. Nucl. Phys. **34**, 662 (1981)].