THE TWO-CENTER PROBLEM IN QUANTUM MECHANICS

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Results of a numerical solution of the two-center problem in quantum mechanics are presented. Some interesting features of the Z_1eZ_2 system are found for $Z_1 = 1$ and $Z_2 = 3, \ldots 8$. In particular, the existence of intersections of σ-terms in such a system is confirmed. These intersections have previously been predicted theoretically [2]. The existence of stable molecular ion states for large distances between the nuclei Z_1 and Z_2 is proved. Pseudointersection of terms, which is not of a dynamic nature, is detected. Possible applications of the results obtained are discussed.

1. INTRODUCTION

THE Schrödinger equation for the two-center problem, that is, the problem of motion of an electron in a field of two fixed Coulomb centers with charges Z_1 and Z_2, separated by a distance R, is [1,2]

$$\left(-\frac{1}{2}\Delta_{\mathbf{r}} - \frac{Z_1}{r_1} - \frac{Z_2}{r_2}\right)\psi_n(R;\mathbf{r}) = E_n(R)\psi_n(R;\mathbf{r}) \quad (1)$$

r_1 and r_2 are the distances from the electron to charges Z_1 and Z_2, respectively; $\hbar = m = e = 1$).

This classical problem of quantum mechanics plays the same role in the theory of the structure and spectrum of molecules as the problem of the hydrogen atom in the theory of atomic structure. Recently, the needs of meson physics at low energies have again renewed the interest in the two-center problem [3,4].

However, in spite of the understandable interest in this problem and the large number of papers devoted to it (see, for example, [1-3]), it has not yet been solved in general form, that is, for arbitrary values of Z_1, Z_2, and R. There are two reasons for it: first, for arbitrary R the problem cannot be solved analytically in terms of the known special functions; second, the exact numerical solution of the problem, obtained in the papers of Bates and his co-workers [5] for $Z_1 = 1$ and $Z_2 = 1$ and 2, meets with peculiar computational difficulties when Z_2 is increased further. The gist of these difficulties and the methods of their overcoming is the subject of a separate mathematical investigation, on which we shall not dwell now [6]. We note also the following: when the variables are separated in spheroidal coordinates

$$\xi = \frac{r_1 + r_2}{R}, \quad \eta = \frac{r_1 - r_2}{R}, \quad \psi = X(\xi)Y(\eta)e^{im\varphi}$$

Eq. (1) reduces to a coupled system of Sturm-Liouville equations on the interval $-1 \le \eta \le 1$, $1 \le \xi < \infty$:

$$\frac{d}{d\xi}(\xi^2 - 1)\frac{dX}{d\xi} + \left[-p^2(\xi^2 - 1) + b'\xi + \lambda - \frac{m^2}{\xi^2 - 1}\right]X = 0, \quad (2a)$$

$$\frac{d}{d\eta}(1 - \eta^2)\frac{dY}{d\eta} + \left[-p^2(1 - \eta^2) + b\eta - \lambda - \frac{m^2}{1 - \eta^2}\right]Y = 0, \quad (2b)$$

where

$$p^2 = -\frac{R^2}{2}E, \quad b' = R(Z_2 + Z_1), \quad b = R(Z_2 - Z_1),$$

λ is the separation constant. With this, Eqs. (2a) and (2b) are coupled only via the quantities p^2 and λ. By solving (2a) and (2b) simultaneously, we determine the eigenvalues $E = E_{n_\xi n_\eta m}(R)$ and $\lambda = \lambda_{n_\xi n_\eta m}(R)$ as functions of the internuclear distance R and of the elliptic quantum numbers n_ξ, n_η, and m. With so many parameters, the tabulation of the functions and of the eigenvalues is difficult. It is apparently likewise inexpedient, for the same reason, to compile extensive tables for all these quantities: at the present status of computational mathematics it is much more convenient to develop a computer program for their determination.

A detailed description of such an algorithm for the calculation of the eigenvalues and the eigenfunctions of (2), and also of a procedure for cal-

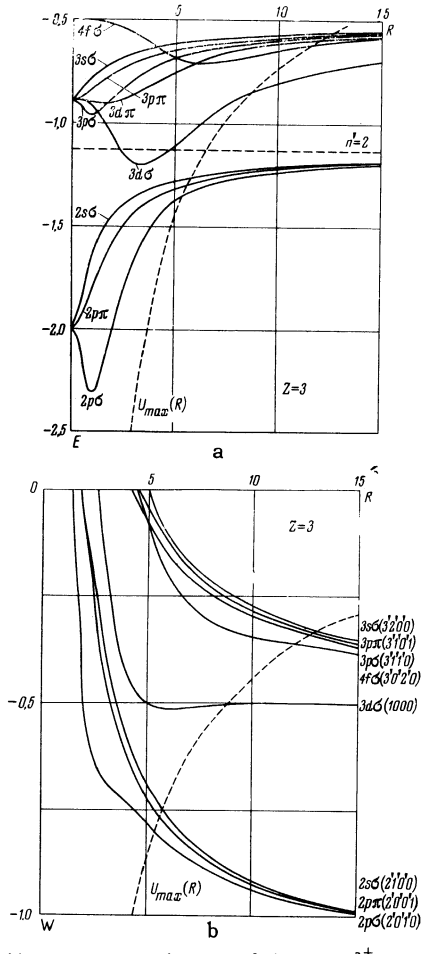

FIG. 1. Terms E(R) and W(R) of the $peLi^{3+}$ system. The $U_{max}(R)$ curve corresponds to the height of the potential barrier between the nuclei.

culating the matrix elements by means of these functions, will be subject of a separate mathematical paper [6].

In this paper we dwell only on the calculation results that have a direct physical interest.

2. THE TERMS E(R) OF THE SYSTEMS $peLi^{3+}$ peB^{5+}, AND peO^{8+}

Usually, for most qualitative considerations, great interest attaches to knowledge of the E(R) terms of the Z_1eZ_2 system. Figures 1a, 2a, and 3a show pictures of the terms $E_{n_\xi n_\eta m}(R)$ for Z_1eZ_2 systems with $Z_1 = 1 = p$ and $Z_2 = Z = 3$, 5, and 8 respectively. Figures 1b, 2b, and 3b show also plots of the total energy $W(R) = E(R) + Z_1Z_2/R$, which includes the nuclear repulsion energy. The classification of the terms is based on the quantum numbers Nlm of the combined atom with charge $Z_1 + Z_2$ into which the system Z_1eZ_2 goes over when $R \to 0$. With this, each level with principal quantum number N is N^2-fold degenerate. When $R \neq 0$ the degeneracy is

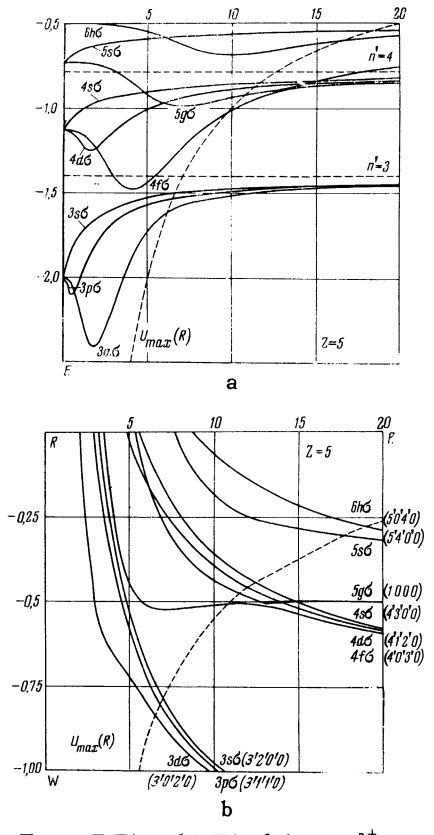

FIG. 2. Terms E(R) and W(R) of the peB^{3+} system. We call attention to the unique interaction of the terms $5g\sigma$ and $4f\sigma$.

lifted and in the limit as $R \to \infty$ each of the levels of the combined atom $e(Z_1 + Z_2)$ goes over continuously into one of the levels of the atom eZ_1 or eZ_2 with parabolic quantum numbers (nn_1n_2m) or $(n'n_1'n_2'm)$ respectively. The relations of these sets of quantum numbers with each other and also with the elliptic quantum numbers N, N_ξ, n_η, and m are given in a paper by Gershteĭn and Krivchenkov [2]. In Figs. 1b—3b the parabolic quantum numbers are given in the parentheses on the right sides of the term symbols.

Let us analyze the obtained curves. First, it follows from them immediately that in this case the Neumann-Wigner theorem, which states that terms of identical symmetry do not cross, does not hold true (in its usual formulation for the case of a diatomic molecule [7]). This result confirms to the theoretical conclusions of Gershteĭn and Krivchenkov [2] that this theorem is not applicable in the case of equations with separable variables.

Actually, however, the presence of such crossings does not contradict the general formulation of the Neumann-Wigner theorem: as shown in a recent paper by Alliluev and Matveenko [8], the problem with separable variables can have a symmetry higher than geometric. From the point of view of this higher symmetry, all the terms of

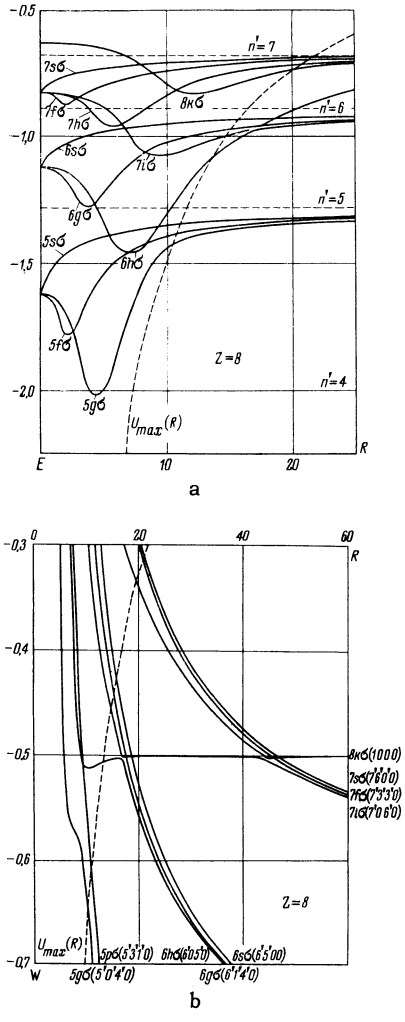

FIG. 3. Terms E(R) and W(R) of the system peO^{8+}. The terms 8kσ, 7iσ, and 6hσ interact.

the system Ze_1eZ_2 have different symmetries and can therefore intersect without limitations. In particular, the σ-terms intersect both for equal N (for example terms 4pσ and 4fσ for Z = 5) and for different N (the terms 5gσ and 4dσ for Z = 5; henceforth $Z_1 = 1$ and $Z_2 = Z$ throughout). The $U_{max}(R)$ curve of Figs. 1a—3a[1)] corresponds to the maximum potential energy U of the electron in the field of two fixed nuclei (that is, to the height of the potential barrier between the nuclei p and Z)

$$U = \frac{1}{r_1} - \frac{Z}{r_2}$$

$$U_{max}(R) = -\frac{(1+\sqrt{Z})^2}{R}. \qquad (3)$$

When $R \gg 1$, the energy of the terms of the peZ system, corresponding when $R \to \infty$ to the levels of the atom ep, is equal to [2]

$$E_n \approx -\frac{1}{2n^2} - \frac{Z}{R}. \qquad (4)$$

Equating expressions (3) and (4), we find that when

$$R = R_x = 2n^2(1 + 2\sqrt{Z}) \qquad (5)$$

the energy of the system peZ becomes equal to the height of the potential barrier between the nuclei p and Z [4], with

$$E_n(R_x) = U_{max}(R_x) = -(1+\sqrt{Z})^2 / 2n^2(1+2\sqrt{Z}).$$

At the point R_x where the term $E_n(R)$ crosses the $U_{max}(R)$ curve, the level n of the isolated hydrogen atom ep goes over into the general level of the system peZ. This phenomenon is perfectly analogous to the vanishing of the Stark-effect lines in strong fields[2)]. Formula (5) was derived under the assumption that the LCAO method is valid. Exact calculation [6] shows that, owing to pseudocrossing effects (which will be discussed in detail below), this formula does not hold, and actually $R_x > 2n^2(1 + 2\sqrt{Z})$ (see Figs. 1a—3a).

3. STABLE STATES OF THE MOLECULAR IONS peZ

The term with quantum numbers n = 1 and $n_1 = n_2 = m = 0$, which corresponds when $R \to \infty$ to the ground state of the hydrogen atom ep, goes over into the terms with quantum numbers N = Z, $l = Z - 1$, and m = 0 when $R \to 0$ [2]. The energy of these terms depends on R in the following manner [2]:

$$\text{when } R \gg 1 \quad W \approx -\frac{1}{2} - \frac{9}{4}\frac{Z^2}{R^4}, \qquad (6a)$$

$$\text{when } R \ll 1 \quad W \approx -\frac{(Z+1)^2}{2Z^2} + \frac{Z}{R}, \qquad (6b)$$

That is, these terms should have a minimum at finite values $R = R_0$ [10]. Actually, as seen from Figs. 1b—3b, the corresponding minima for the terms 3dσ with Z = 3, 5gσ with Z = 5, and 8kσ with Z = 8 are attained at internuclear distances R_0 equal to 6.2, 13.6, and 44.5. This means that at large internuclear distances there exist stable states of the molecular ions of the peZ type.

It has turned out, however, that in addition to the states of the molecular ions, quasistationary

[1)] The U_{max} (R) curves on the plots of W(R) (Figs. 1b – 3b) differ from the U_{max} (R) curves on the plots of E(R) by a term Z/R.

[2)] The critical field F at which the line vanishes can be easily obtained from (3) and (5):

$$F = (1+\sqrt{Z})^2 / 4n^4(1+2\sqrt{Z})^2.$$

When $Z \to \infty$ we obtain the classical limit [9]: $F_0 = 1/16n^4$.

levels are possible in peZ system. They appear when $Z > 4$ and are connected with the terms $N = Z - 1$, $l = Z - 2$, and $m = 0$ (that is, $n' = Z - 1$, $n_1' = 0$, $n_2' = Z - 2$, and $m = 0$; for example, $4f\sigma$ for $Z = 5$, and $7i\sigma$ and $6h\sigma$ for $Z = 8$). Apparently this type of terms was hitherto unknown.

4. TERM PSEUDOINTERSECTION PHENOMENA

We call attention to the unique interaction of the terms with the quantum numbers

$$N = Z, \quad l = Z - 1, \quad m = 0$$

$$(n = 1, \quad n_1 = n_2 = m = 0)$$

and

$$N = Z - 1, \quad l = Z - 2, \quad m = 0$$

$$(n' = Z - 1, \quad n_1' = 0, \quad n_2' = Z - 2, \quad m = 0).$$

This phenomenon occurs when $Z \geq 4$, although it is noticeable already when $Z = 3$ for the terms $3d\sigma$ and $2p\sigma$ (see Fig. 1b). With increasing Z, these terms come gradually closer together, forming at the same time a typical picture of pseudointersection (for example, the terms $5g\sigma$ and $4f\sigma$ for $Z = 5$ on Fig. 2 and $8k\sigma$ and $7i\sigma$ for $Z = 8$ on Fig. 3).

When $Z \geq 7$, the pseudointersection picture is produced by one more pair of terms

$$N = Z - 1, \quad l = Z - 2, \quad m = 0$$

$$(n' = Z - 1, \quad n_1' = 0, \quad n_2' = Z - 2, \quad m = 0)$$

and

$$N = Z - 2, \quad l = Z - 3, \quad m = 0$$

$$(n' = Z - 2, \quad n_1' = 0, \quad n_2' = Z - 3, \quad m = 0)$$

(for example, $7i\sigma$ and $6h\sigma$ for $Z = 8$ in Fig. 3). With increasing Z, this tendency still remains, that is, the number of such "interacting" pairs of terms increases.

Let us note the characteristic attributes of the described terms: all have a maximum angular momentum $l = N - 1$, which corresponds to circular orbits in Bohr's theory and to the largest density of the electron cloud between the nuclear Z_1 and Z_2 in the Stark effect, for in this case $n_2 - n_1$ is a maximum [9]; in addition, their "radial" wave function $X(\xi)$ does not have any zeroes in the entire region where ξ is defined, that is, $n_\xi = n_1 = n_1' = 0$, and the "angular" quantum numbers differ by unity: $n' = n + 1$.

We note one more peculiarity of these pseudointersections: for the same value of R as for W(R), a pseudointersection of the separation

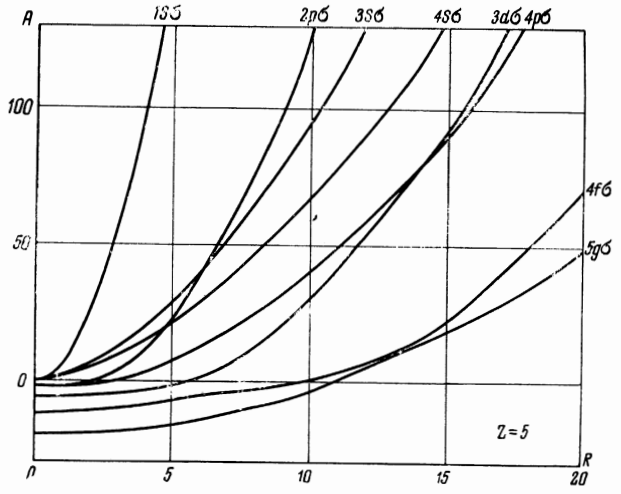

FIG. 4. Values of $A = \lambda + p^2$ for different terms at $Z = 5$.

constants $\lambda(R)$ of the corresponding levels takes place for them, whereas for the ordinary intersections the constants $\lambda(R)$ differ greatly at the point of the term crossing W(R) (see Fig. 4, which gives the values of $A = \lambda + p^2$ for different terms at $Z = 5$). We note that by virtue of the nondegeneracy of the one-dimensional boundary-value problem, the $\lambda(R)$ and W(R) curves for different levels cannot intersect at the same value of R [2].

The unique "interactions" of the terms are also revealed by the fact that in the pseudointersection region the term with $N = Z - 1$ has the properties of the term with $N = Z$. In particular, for the term with $N = Z$ ($5g\sigma$ for $Z = 5$), in the LCAO approximation, at values of R to the right of the point of pseudo-intersection, formula (6a) holds for the polarization of the ground level of the hydrogen atom, into which the term $5g\sigma$ goes over as $R \to \infty$. It turns out that to the left of the pseudointersection point formula (6a) describes well the term with $N = Z - 1$ ($4f\sigma$ for $Z = 5$), that is, formally the term $5g\sigma$ and $4f\sigma$ intersect. Actually, of course, this does not take place, since it follows from an analysis of the behavior of the terms as $R \to 0$ (see Fig. 2a) that in this case the terms $5g\sigma$ and $4f\sigma$ do not simply intersect but go over one into the other. The latter is impossible, for this is equivalent to violation of the theorem regarding the conservation of the number of zeroes of the wave function $Y(\eta)$ when the parameter R varies, and violation of such corollaries of theorem as the rule of correspondence of the terms for $R \to 0$ and $R \to \infty$ [2].

Gershteĭn, using the LCAO method, obtained the usual intersection of the terms at the point of pseudointersection, meaning that the LCAO method is not valid in this range of values of R.

This is all the more strange, since all the indicated pseudointersections occur either in the region of the potential barrier ($E(R) \approx U_{max}(R)$) or behind the barrier, that is, when $E(R) < U_{max}(R)$ (see Figs. 2a and 3a).

This gives rise to a unique situation: The LCAO formulas, which are well satisfied in a wide range of values of R on both sides of the pseudointersection point, are quite unsuitable in the vicinity of this point, since they lead to the crossing of the terms. According to the results of Gershteĭn and Krivchenkov [2] and Alliluev and Matveenko, and also according to Smirnov [8], there are no general hindrances for any crossings of the terms in Z_1eZ_2 system, so that additional selection rules, which distinguish the terms with $n_1 = n_1' = 0$ from all others, should exist in this case.

Let us examine this phenomenon in greater detail[3]. When $R \to \infty$ the LCAO method leads to the following expressions for the total energy $W(R)$ of the peZ system:

$$W_1 \approx -\frac{1}{2} - \frac{9}{4}\frac{Z^2}{R^4} \qquad (N = Z,\ l = Z-1),$$

$$W_2 \approx -\frac{1}{2}\left(\frac{Z}{Z-1}\right)^2 + \frac{Z-1}{R} - \frac{3}{2}\frac{(Z-1)(Z-2)}{ZR^2} \qquad (N = Z-1,\quad l = Z-2). \tag{7}$$

We then obtain from the condition $W_1 = W_2$ an intersection of the terms at the point $R = R_Z$

$$R_Z \approx 2\frac{(Z-1)^3}{2Z-1}. \tag{8}$$

However, from (2a) it follows that for any R there should be satisfied the relation [2] ($A = \lambda + p^2$):

$$(A_1 - A_2)\int_1^\infty X_1X_2d\xi = (p_1^2 - p_2^2)\int_1^\infty X_1\xi^2X_2d\xi. \tag{9}$$

Therefore we get from the condition $p_1 = p_2$ at the intersection point either that $A_1 = A_2$ or that $\int_0^\infty X_1X_2d\xi = 0$. The former is impossible, since the problem is nondegenerate. For terms with quantum numbers $n_1 = n_1' = 0$ the latter condition is also impossible, for in this case the integrand is positive in the entire defined region. Consequently, such terms cannot intersect, although they can come close to one another. It follows from (9) that when $\Delta p^2 = p_1^2 - p_2^2 \to 0$ we have simultaneously $\Delta A \to 0$. The latter can also be established directly, by using the expressions given in [2] for A when $R \gg 1$ and $n_1 = n_1' = 0$.

In this case

$$\frac{\Delta A}{\Delta p^2} = 1 + \frac{1}{p} + O\left(\frac{1}{p^2}\right). \tag{10}$$

We note here one quasiclassical analogy[4], which may be useful for the understanding of the described phenomenon. As is well known [11], when $Z_1 = Z_2$ the effective potential of Eq. (2b) has the form of a double symmetrical well. In this case, when $R \to \infty$, the levels with the quantum numbers n_ξ, n_η, m and $n_\xi' = n_\xi$, $n_\eta' = n_\eta + 1$, m are degenerate, that is, they have equal energy. For a large but finite value of R, the splitting of these levels is exponentially small and is described, for example, by formula (23) of [3]. In the general case when $Z_1 \neq Z_2$, this double well is asymmetrical, but its concrete form depends on the parameter R. For a certain value of R (but a value below the barrier!), which depends on Z_1 and Z_2 and also on the quantum numbers of the levels, the effective potential of (2b) may turn out to be such that the two levels in different wells approximately coincide. In this case, phenomena analogous to the effects of the level splitting in a symmetrical potential should occur. More accurately speaking, the levels whose energies are not equal when $R \to \infty$, and whose wave functions are concentrated in different wells, can come closer together when R decreases and start "interacting" as a result of quantum-mechanical subbarrier transitions. With further change in R, this random "resonance" of the levels is disturbed and they again move apart.

We emphasize that the observed "interaction" of the terms has no dynamic nature, since the system peZ has only one electron, and the nuclei are assumed to be fixed[5]. We have here essentially an example of a unique interaction of configurations, and therefore the described phenomenon can be called configurational interaction of the terms.

5. WAVE FUNCTIONS OF THE peZ SYSTEM

Figures 5 and 6 show the wave functions $X(\xi)$ and $Y(\eta)$ of the peZ system for $Z = 5$ and for the terms $5g\sigma$, $4f\sigma$, and $4s\sigma$. As expected, as $R \to 0$ they describe the motion of an electron in the field of a nucleus with charge $Z + 1$.

[3] These ideas are due to S. S. Gershteĭn.

[4] This circumstance was pointed out to the authors by S. P. Alliluev.

[5] Usually the pseudo-intersection picture arises at the point of level intersection when account is taken of perturbations due to motion of the nuclei, electron intersection, etc. [7].

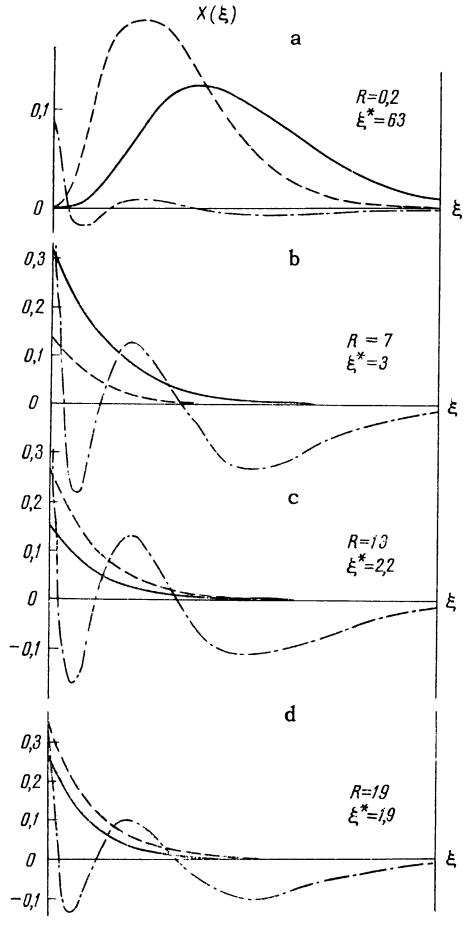

FIG. 5. Wave functions $X(\xi)$ of the peZ system with Z = 5 for the terms $5g\sigma$ (solid curve), $4f\sigma$ (dashed), and $4s\sigma$ (dash-dot). The functions $X(\xi)$ are not normalized. At R = 0.2 the function $X(\xi)$ for the $4s\sigma$ term is decreased by a factor of 10; ξ^* is determined from the relation $\xi^* = 10/p + 1$, $p = 1/2R\sqrt{-2E}$. The abscissa scales are different for different R and depend on ξ^*, the values of $X(\xi)$ being defined in the interval $1 \leq \xi \leq \xi^*$.

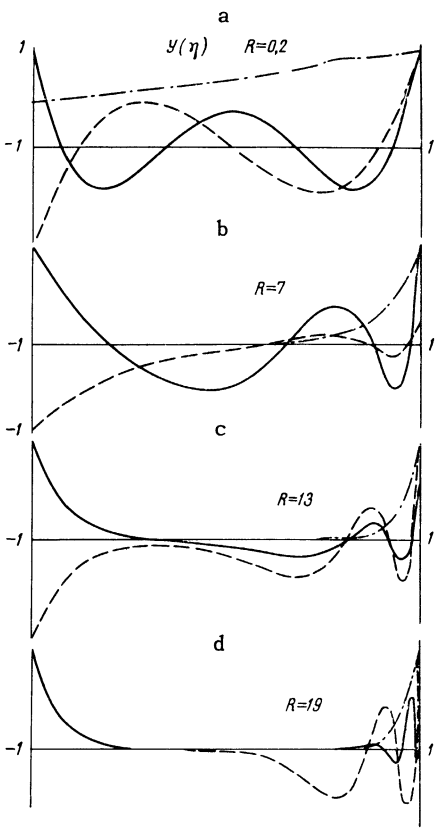

FIG. 6. Wave functions $Y(\eta)$ of the peZ system with Z = 5 for the terms $5g\sigma$ (solid curve), $4f\sigma$ (dashed), and $4s\sigma$ (dash-dot).

We call attention to the peculiarities of the behavior of the wave functions at R = 7 and R = 13. In particular, when R = 13 the wave functions of the terms $5g\sigma$ and $4f\sigma$ are quite similar. Further, in spite of the fact that when $R \to \infty$ the term $4f\sigma$ belongs to the eZ system, its wave function is already concentrated near the proton when R = 7. This corresponds precisely to the statement that the term $4f\sigma$ acquires the properties of the term $5g\sigma$ to the left of the pseudo-intersection point. With further increase in R, the wave functions of the peZ system go over on the right of the pseudo-intersection point (R = 19) into the wave functions of the isolated atoms ep and eZ, as can be readily seen from Figs. 5d and 6d.

6. POSSIBLE APPLICATIONS

1. Using the results of the present paper, we can estimate the character and limits of applicability of different approximations, and particularly the values of R for which the asymptotic formulas are valid. For example, from an analysis of the curves it follows that when $R \gg 1$ there exists a region of values of R where the LCAO approximation is not valid.

2. Our results allow us to calculate the probabilities of radiative transitions of a π^- meson moving in the Coulomb field of two fixed nucleons. Such a calculation is of physical interest in connection with experimental [12] and theoretical [4] research on the absorption of π^- mesons stopped in hydrogen-containing substances, and also in connection with experiments on the mesic x-ray series in chemical compounds.[13]

3. The theory of the chemical bond and of molecular spectra makes extensive use of the method of molecular orbitals [3], which are usually determined approximately by using linear combinations of atomic functions in conjunction with a

variational principle. It is obvious that by solving (1) we obtain an exact picture of the molecular orbitals.

4. In some estimates of the cross sections of charge-exchange reactions of the type $A + B^+ \rightarrow A^+ + B$ and their like it is sufficient to know the distance R at which the intersection of the terms takes place, as well as the values of the derivatives $\partial E/\partial R$ at this point.

All these quantities can be easily determined if the system of terms is known.

5. The main application of our results, however, is connected with various processes in which three Coulomb-interacting particles participate. These include, in particular, problems of capture and asymmetrical charge exchange of the type

$$p\mu^- + Z \rightarrow Z\mu^- + p,$$

$$p\mu^- + Z \rightarrow Z\mu^- + p + \gamma;$$

mesic-molecule production processes

$$p\mu^- + Z \rightarrow p\mu^- Z;$$

catalysis of nuclear reactions by μ^- mesons

$$p\mu^- + Z \rightarrow p\mu^- Z \rightarrow (Z+1) + \mu^-;$$

scattering and protons and positrons by hydrogen atoms, and similar questions.

In conclusion, we are deeply grateful to S. S. Gershteĭn for persistent and stimulating interest during the course of the entire investigation, and S. P. Alliuev for fruitful discussions.

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