Interaction of an excited atom with an atom of small radius

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(Submitted March 4, 1974)
Zh. Eksp. Teor. Fiz. 68, 12-21 (January 1975)

The interaction between an excited atom and a potential well of small radius is discussed. Expressions valid for an arbitrary atom are obtained for the asymptotic energy shifts of the atomic and ionic energy levels in the region of their quasi-intersection. It is shown that in the purely Coulomb case the splitting of terms on quasi-intersection as a result of the degeneracy of the levels is approximately $\sqrt{m}$ times greater than for an arbitrary atom with the same energy (m is the principal quantum number). It is also shown that all of the asymptotic formulas have higher accuracy (for expansion in inverse powers of the internuclear distance) than was assumed previously.

1. The interaction between a bound electron which has an extended wave function and a potential well of small dimensions has been discussed in many theoretical papers, which have been collected, for example, in the book by Smirnov.[1] The main result of these studies is that the electron energy shift from the initial unperturbed value E_m is

$$E(R)-E_m=2\pi L|\psi_m(R)|^2, \tag{1}$$

where L is the scattering length of the electron in the unperturbed potential well, ψ_m is the wave function of the initial unperturbed state of the electron in the atom interacting with the well considered, and R is the distance from the nucleus of the atom to the center of the perturbing well, which is assumed spherically symmetric. The atomic system of units is used: $e = m = \hbar = 1$.

The result (1) is the zeroth approximation in the expansion of the exact wave function in a small parameter which is the ratio of the extent of the perturbing well to the electron wavelength. The method of obtaining this result is based on matching the exact wave function of the electron, which satisfies the wave equation

$$[-\Delta/2+V_1(r)+V_2(|\mathbf{r}-\mathbf{R}|)-E]\Psi(r)=0, \tag{2}$$

with the unperturbed wave function

$$[-\Delta/2+V_1(r)-E_m]\psi_m(r)=0. \tag{3}$$

Here V_1 is the interaction of the electron with its atomic core, and V_2 is the interaction with the perturbing well. Since the size of the region of perturbation of the exact wave function is L, the expansion parameter in this solution of the problem will be $L(-2E_m)^{1/2} \ll 1$, and also $r_2(-2E_m)^{1/2}$ (r_2 is the size of the well V_2), for it is necessary that at distances $\sim L$, r_2 from the center of the well V_2 the function ψ_m not vary strongly. It is easy to see that this construction of the solution will be valid only for energy shifts small in comparison with the distance between the atomic levels, since otherwise the solution beyond the limits of the perturbation region must differ greatly from ψ_m. On the other hand, Eq. (1) already shows that the energy shifts can be large. In fact, if the perturbing well falls in the region of the main distribution of electron density, then here the square of the modulus of the wave function is $\sim m^{-3}$, i.e., of the order of the distance between levels.

Another method of determining the electronic levels proposed by Komarov et al.[2,3] does not require fulfillment of the first of the above criteria, since the calculations are carried out with use of a source function for the main atomic field. Since the perturbing objects are neutral atoms whose extent is determined by their polarizability α, this method works for the condition

$$(2\alpha|E_m|)^{1/2}\ll 1. \tag{4}$$

According to Komarov et al.[2,3] the energy levels are determined from the condition for the logarithmic derivative of the electronic Green's function G of the field V_1:

$$\left[\frac{\partial}{\partial\rho}\rho G(\mathbf{r},\mathbf{R},E)\right]_{\rho\to 0}\equiv\Phi(E,R)=-\frac{1}{L}, \tag{5}$$

$\rho \equiv |\mathbf{r}-\mathbf{R}|$. This condition is obtained in the zeroth approximation in expansion of the solution of Eq. (2) in the small parameter (4), which corresponds to replacement of the well V_2 by an effective δ-function well with a scattering length L.

Komarov et al.[2,3] investigated Eq. (5) for the purely Coulomb function G obtained by Hostler et al.[4,5] In this work the energy levels given by Eq. (5) are studied for the case of an arbitrary atom in which the field V_1, in addition to the asymptotic Coulomb part, has an added core. It is shown that because of the specific degeneracy of the levels the purely Coulomb case is special.

2. We will investigate the asymptotic shift of the levels when the perturbing well V_2 is far in the subbarrier region (for V_1) of motion of the electron. We assume that $V_1 \to -r^{-1}$ as $r \to \infty$ and has a core for $r \sim 0$. Since, as the perturbing well is made more remote, the levels approach the unperturbed levels of the wells V_1 and V_2, it is necessary to find a double expansion of the function $G(\mathbf{r},\mathbf{R},E)$ in inverse powers of R and powers of the energy shift $E - E_m$.

As shown in the next section, the function G for $R \gg (-2E)^{-1}$ can be written in the form

$$\begin{gathered}G(\mathbf{r},\mathbf{R},E)=\frac{\psi_m(\mathbf{r})\psi_m(\mathbf{R})}{E_m-E}+\frac{\overline{W}'(y/n)W(x/n)-\overline{W}(y/n)W'(x/n)}{2\pi|\mathbf{R}-\mathbf{r}|}\\+O(R^{-4})+O(|\psi_m(R)|^2),\quad n\equiv(-2E)^{-1/2},\\x\equiv r+R+|\mathbf{R}-\mathbf{r}|,\quad y\equiv r+R-|\mathbf{R}-\mathbf{r}|.\end{gathered} \tag{6}$$

Determining the quantity $\Phi(E,R)$ by expanding of the Whittaker functions W in Eq. (6) (see Eq. (28) below) and substituting it into Eq. (5), we obtain an equation for determination of the energy:

$$\frac{2\pi|\psi_m(R)|^2}{E_m-E}+\frac{1}{L}-\left(-2E-\frac{2}{R}\right)^{1/2}=0. \tag{7}$$

If the electron with well V_2 forms ($L > 0$) a bound state (negative ion) with an energy not falling in resonance with the atomic levels, we find the behavior of this ionic level, discarding the first term from Eq. (7):

$$E_-(R)=E_-(\infty)-1/R,\quad E_-(\infty)=-1/2L^2. \tag{8}$$

As we see, retention of the radical in Eq. (7) and (8) in a form not expanded in powers of R^{-1} is convenient for a determination of the ionic level, although it is actually exceeding the accuracy. This excess, however, occurs in a remote order proportional to R^{-4} (see Eq. (28)).

In the region where $E_-(R)$ is close to the atomic levels, Eq. (8) is not valid, since in this case the first term in Eq. (7) cannot be discarded. For a correct determination of the point of intersection of the ionic level with the atomic level, we retain the radical in Eq. (7) in the unexpanded form as before. Then Eq. (7) is easily reduced to a cubic equation, one root of which is superfluous:

$$(E-E_m)^3+[E_m-E_-(R)](E-E_m)^2-\frac{2\pi|\psi_m(R)|^2}{L}(E-E_m)+2\pi^2|\psi_m(R)|^4=0. \tag{9}$$

Here $E_-(R)$ is determined by Eq. (8). At the point of intersection the coefficient in the quadratic term goes to zero. Near this point in Eq. (9) we can neglect the free term, which leads to a solution

$$E=\frac{E_m+E_-(R)}{2}\pm\frac{1}{2}\left[(E_m-E_-(R))^2+\frac{8\pi}{L}|\psi_m(R)|^2\right]^{1/2}. \tag{10}$$

In this way there is a quasi-intersection of the atomic and ionic terms with a minimum distance between them of

$$\min|\Delta E|=(8\pi/L)|\psi_m(R_0)|,\quad E_m-E_-(\infty)=-1/R_0. \tag{11}$$

This result is valid for intersection of the ionic term with each of the atomic terms.

Equation (10) agrees with the well known expression for the splitting of terms in the two-level approximation (as should be the case), the nondiagonal matrix element being a factor of two smaller than expression (11). Far from the point of intersection, Eq. (10) describes the shift of the atomic terms incorrectly, since in this region one cannot neglect the free term in Eq. (9). The correct shift of the atomic terms far from the point R_0 is found by setting $E = E_m$ under the radical sign in Eq. (7):

$$E=E_m+\frac{2\pi L|\psi_m(R)|^2[1+O(R^{-4})]}{1-L[-2E_m+2V_1(R)]^{1/2}}. \tag{12}$$

This expression agrees with the result of Ovchinnikova[6] except for the estimate of the accuracy. For $R > R_0$ the denominator here is less than zero and we obtain an attraction between the wells ($L > 0$), and for $R < R_0$ in a repulsion. At the point R_0 itself, instead of Eq. (12) it is necessary to use Eq. (10). For a negative scattering length ($L < 0$), Eq. (12) is always applicable in the sub-barrier region. In Eq. (12) both the radical and the square of the modulus of the wave function must be taken with an accuracy to R^{-4}, inclusive. To the left of the classical turning point the radical in Eq. (12) becomes imaginary. Then we can utilize an expansion of the radical and obtain

$$E=E_m-\frac{2\pi L|\psi_m(R)|^2}{1-L(-2E_m)^{1/2}-LV_1(R)/(-2E_m)^{1/2}}. \tag{12a}$$

Equation (12) describes also the case of initial resonance, where $E_m = E_-(\infty)$. The shift of the atomic level for this case is

$$E=E_m-\frac{2\pi(-2E_m)^{1/2}}{V_1(R)}|\psi_m(R)|^2. \tag{13}$$

It is evident that the resonance situation for $R = \infty$ is in fact weakly resonant, since the rapid shift of the ionic level (which always is given by Eq. (8)) spoils the resonance. The true resonance situation appears on intersection of the ionic and atomic levels. Only in this case do we have an appearance of the exchange-charge-transfer interaction (which is proportional to expression (11)), due to a sub-barrier transition of an electron from one well to the other.

In Smirnov's work[7] the separation of the terms on quasi-intersection of the atomic and ionic states was studied by a method different from that presented here. Specifically, the wave function of the ionic state with inclusion of the Coulomb field was determined in the quasiclassical approximation with boundary conditions specified at the ion boundary. The result was

$$\begin{gathered}\Psi_{\text{ion}}\approx A\sqrt{\frac{2}{n}}\frac{e^{-\rho/n}}{\rho}\chi(\rho)\left[1+O\left(\frac{1}{R}\right)\right],\\ \chi(\rho)=\left(\frac{R}{R-\rho}\right)^n e^{-n\rho/R};\quad \mathbf{r}\|\mathbf{R},\quad r<R.\end{gathered} \tag{14}$$

Here A is a coefficient that accounts for the correct amplitude of the unperturbed ionic function at large distances ($A = 1$ in the model $V_2 \approx \delta(\rho)$). The method of obtaining the Coulomb Green's function used in ref. 4 permits an exact expression to be written for the ionic function. It is identical with the second term in Eq. (6), so that (see Eq. (22))

$$\begin{gathered}\Psi_{\text{ion}}\approx N\frac{\overline{W}'(y/n)W(x/n)-\overline{W}(y/n)W'(x/n)}{|\mathbf{R}-\mathbf{r}|}\approx N\frac{e^{-\rho/n}}{\rho}\left(\frac{R}{R-\rho}\right)^n;\\ r\sim R\gg n^2,\quad \mathbf{r}\|\mathbf{R},\quad r<R.\end{gathered} \tag{14a}$$

As we see, the exact solution (14a) does not contain the weak quasi-classical exponential $e^{-n\rho/R}$, as a result of which the expression obtained by Smirnov[7] for the splitting of the terms on quasi-intersection differs from Eq. (11) by a factor $[mL/re^m]^{1/2}$. The question of improving the quasiclassical solution is fundamental in nature and remains open.

Thus, the method discussed here permits a single description of all particular cases with substantial improvements in the results. In addition, the asymptotic formulas (10)–(13) written out above are suitable for any potential V_1. Thus, the asymptotic behavior of the terms for the case discussed by Smirnov[8] of two δ-function wells is obtained from these formulas if we set $V_1 \equiv 0$.

3. Before deriving and investigating Eq. (6), we will briefly study the pure Coulomb case, for which there is a closed expression for the Green's function obtained by Hostler[4,5]:

$$G_0(E,\mathbf{r},\mathbf{R})=\frac{\Gamma(1-n)}{2\pi|\mathbf{R}-\mathbf{r}|}\left[M'\left(\frac{y}{n}\right)W\left(\frac{x}{n}\right)-M\left(\frac{y}{n}\right)W'\left(\frac{x}{n}\right)\right], \tag{15}$$

where the variables x and y are defined in Eq. (6); M and W are Whittaker functions, M being regular at the origin and W at infinity.[9] The quantity Φ defined by means of Eq. (15) is listed in ref. 2. For investigation of the asymptotic behavior of the terms it is convenient to use another representation for Φ:

$$\Phi(E,R)=-\frac{1}{n}+2\Gamma(1-n)\int_{2R/n}^{\infty}\frac{M(\xi)W(\xi)}{\xi^2}d\xi, \tag{16}$$

which is obtained by integration of the sum of the Whittaker equations for M and W.

Equations (5) and (16) permit investigation of the shift of the Coulomb (hydrogen) terms in numerical integration of the Whittaker equations. In Figs. 1–3 we give the results of such numerical calculations. In Figs. 1 and 2 we show the shifts of the atomic levels of hydrogen with n = 2 and n = 3, interacting with wells having the scattering lengths of the helium and argon atoms. It should be noted that the polarizability of the argon atom $\alpha_{\mathrm{Ar}} = 11$ at. un. is so large that criterion (4) for the levels n = 2 and 3 is not satisfied, so that the lower curves in Figs. 1 and 2 describe the interaction with other atoms with the same scattering length but with smaller polarizability.

Using the well known representation of the Green's function in the form of a sum over eigenfunctions, it is easy to find that for a pure Coulomb field in the asymptotic formulas (11) and (10)–(13) the sum of the squares of the moduli of the wavefunctions of all degenerate states must enter. Using the asymptotic forms of these functions,[9] it is easy to obtain

$$|\Psi_m(R)|^2 = \sum_{l=0}^{m-1} |\varphi_{ml0}(R)|^2 \approx m|\varphi_{m00}(R)|^2[1+O(1/R)]. \qquad (17)$$

Consequently, for degenerate states the shift of the atomic levels is larger by a factor m, and the splitting of the terms on quasi-intersection is larger by a factor $\sqrt{m}$, than for an isolated state without angular momentum. This result is also obtained explicitly from Eq. (5) with use of the asymptotic values of the Whittaker functions in Eq. (16). In Figs. 1 and 2 the asymptotic shifts of the levels are given with allowance for Eq. (17).

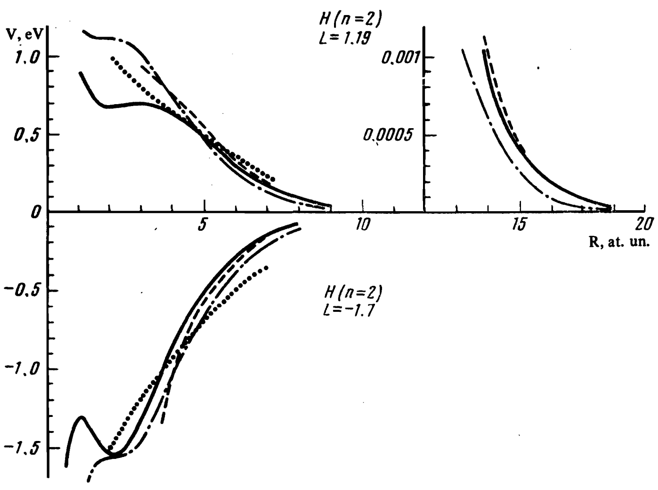

FIG. 1. Interaction of H (n = 2) with atoms having scattering lengths 1.19 (He) and −1.7 (Ar). The solid curve is a numerical calculation, the dot-dash curve is a calculation with Eq. (1), the dashed curve with Eq. (12), and the dotted curve with Eq. (18).

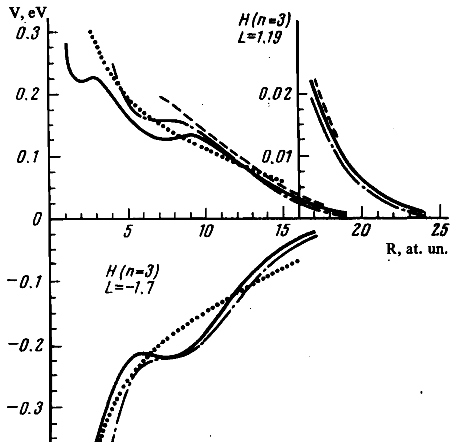

FIG. 2. The same as Fig. 1 but for H (n = 3).

The level splitting obtained in ref. 2 for quasi-intersection for a pure Coulomb field agrees with Eq. (11) with inclusion of relation (17).

In Fig. 3 we show the behavior of the terms for the case L = 3 in the region of quasi-intersection of the ionic level with the atomic level for m = 2. The minimum distance between the terms turned out to be 0.108 eV in the numerical calculation, while Eq. (11) predicts 0.109 eV. The numerical calculation confirms the high accuracy of the asymptotic formulas (10)–(13).

Daladchik and Ivanov[10, 11] have made a study of the terms by means of Eq. (5) for the case in which a perturbing well of small dimensions is in the region of classically allowed motion of the electron in the field V_1. In their articles it is shown that in the region of values R considered the degeneracy of the Coulomb levels affects the splitting of the terms even more strongly. Specifically, the hydrogen terms are shifted according to the relation

$$E-E_m \approx \frac{L}{\pi m^3} p(R), \quad p(R) \equiv \left(\frac{2}{R} - \frac{1}{m^2}\right)^{1/2}. \qquad (18)$$

In Figs. 1 and 2 we have shown the terms given by this formula. As can be seen, the exact (numerical) terms oscillate slightly about the terms (18), which also is predicted in refs. 10 and 11. The difference of the field of real atoms from a Coulomb field is taken into account in refs. 10 and 11 by introduction of a second well of small dimensions placed near the origin, which removes the degeneracy only for the s terms.

4. In the approximation of a potential of zero range, the wave function of the negative ion coincides with the Green's function of the external field in which the negative ion is located. The equality (5) is the condition of matching of this function with the internal part of the wave function. Demkov and Drukarev[12] have used this approximation to study the behavior of the negative ion in a uniform electric field. In the present work this same problem is discussed for a Coulomb field with a

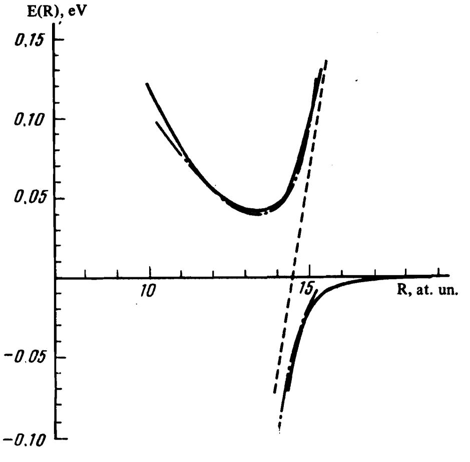

FIG. 3. Region of quasi-interaction of terms for L = 3. Solid curve—numerical calculation, dot-dash curve—calculation with Eq. (10).

core. A feature of the present problem, in comparison with ref. 12, is the existence of bound states and their interaction with the ionic state.

The one-electron Green's function G(**r**,**R**,E) is the solution of the equation

$$\left(-\frac{\Delta}{2}+V_1(r)-E\right)G=\delta(\mathbf{r}-\mathbf{R}). \tag{19}$$

The function (15) is the exact solution of this equation for a pure Coulomb field V_1. Writing the Whittaker function M finite at the origin in the form of combinations of functions $\tilde{W}$ and W having a specified behavior at infinity,[9] we represent (15) in the form

$$G_0^{(1)}=\frac{1}{2\pi|\mathbf{R}-\mathbf{r}|}\left[\tilde{W}'\left(\frac{y}{n}\right)W\left(\frac{x}{n}\right)-\tilde{W}\left(\frac{y}{n}\right)W'\left(\frac{x}{n}\right)\right], \tag{20a}$$

$$G_0^{(2)}=\frac{C(E)\Gamma(1-n)}{2\pi|\mathbf{R}-\mathbf{r}|}\left[W'\left(\frac{y}{n}\right)W\left(\frac{x}{n}\right)-W\left(\frac{y}{n}\right)W'\left(\frac{x}{n}\right)\right], \tag{20b}$$

$$\begin{gathered}M=C(E)W+\frac{\tilde{W}}{\Gamma(1-n)},\quad G_0=G_0^{(1)}+G_0^{(2)},\\ \tilde{W}(\xi)\approx\xi^{-n}e^{\xi/2}\left[1+O\left(\frac{1}{\xi}\right)\right],\quad W(\xi)\approx\xi^{n}e^{-\xi/2}\left[1+O\left(\frac{1}{\xi}\right)\right].\end{gathered} \tag{21}$$

According to the method of constructing G_0 proposed in ref. 4, the function (20a) is an exact particular solution of the inhomogeneous equation (19), and (20b) is the general solution of the homogeneous equation. Using asymptotic expansion[9] of the functions $\tilde{W}$ and W, we obtain an expansion for $G_0^{(1)}$:

$$G_0^{(1)}\approx\frac{e^{-\rho/n}}{2\pi\rho}\left(\frac{r+R+\rho}{r+R-\rho}\right)^n\left[1+\frac{n^3\rho}{rR+\mathbf{rR}}+\ldots\right],\quad r\sim R\gg n^2. \tag{22}$$

We will seek a solution of Eq. (19) for a general potential V_1 (Coulomb plus core) in the form of the sum of a particular solution of the inhomogeneous equation $G^{(1)}$ and the general solution of the homogeneous equation $G^{(2)}$. Since at large $r\sim R\gg 1$ the field V_1 differs from the Coulomb field by an amount $\sim r^{-4}$ determined by the polarizability of the core, $G^{(1)}$ coincides with the same accuracy with $G_0^{(1)}$, where the asymptotic expansion of $G^{(1)}$ is given by the series (22), in which two additional terms can be taken into account.

The second component of the Green's function $G^{(2)}$ should differ substantially from the Coulomb component $G_0^{(2)}$, since the spectrum of its energy poles differs substantially from the Coulomb spectrum of energy eigenvalues.

We seek a solution of the homogeneous equation (19) in the form

$$G^{(2)}=\frac{\psi_m(r)\psi_m(R)}{E_m-E}+\psi_m(r)\psi_m(R)F(r,R), \tag{23}$$

where F is a new unknown function. Substitution of (23) into the homogeneous equation (19) leads to an inhomogeneous equation for F:

$$\left[-\frac{\Delta}{2}-\frac{\nabla\psi_m}{\psi_m}\nabla-(E-E_m)\right]F=-1. \tag{24}$$

We construct a solution of Eq. (23) regular at $r\to\infty$. In the region of large $r\sim R\gg 1$ we can discard from this equation the second derivatives, and in the product of the gradients leave only the derivative in the direction of exponential damping of ψ_m. Then from (24) we arrive at the equation

$$\left[(-2E_m)^{1/2}\frac{\partial}{\partial r}-(E-E_m)\right]F=-1. \tag{25}$$

The solution of this equation symmetric with respect to the substitution $r\rightleftarrows R$ has the form

$$F=\frac{1}{E_m-E}\left[\exp\left\{\frac{E-E_m}{\sqrt{-2E_m}}(r+R)\right\}-1\right]. \tag{26}$$

After obtaining this simple solution, it is evident that the basis for the transition from the exact equation (24) to the approximate equation (25) is the fact that the action of the second derivatives on F leads to appearance of terms of higher order in powers of $E-E_m$, i.e., expression (26) is the principal order of expansion of the function F in powers of $E-E_m$. According to Eqs. (11)–(13), the energy shift $E-E_m$ drops exponentially with increasing R, and therefore

$$\left|\frac{E-E_m}{(-2E_m)^{1/2}}\right|(r+R)\ll 1.$$

Consequently, in the region of values $r\sim R$ the exponential in (26) is not greatly different from unity and therefore

$$F\sim-(r+R)/(-2E_m)^{1/2},\quad r\sim R.$$

As r goes to infinity, i.e., for $r\gg R$, expression (26) increases exponentially for $E>E_m$ and falls exponentially if $E<E_m$. In both cases the complete expression (23) is exponentially damped as $r\to\infty$, since $|E-E_m|\ll|E_m|$.

Thus, we see that the second term in Eq. (23) is a quantity of higher order than the first (exponentially), and can be discarded. Adding (23) and (20a), we arrive at Eq. (6).

The two components of the source function $G^{(1)}$ and $G^{(2)}$ have a singularity at the origin, while their sum—the complete function G—for an accurate solution of Eq. (19) should not have this singularity.

We now find the desired expansion of the logarithmic derivative of the Green's function. By analogy with Eq. (16) and using Eq. (6), we have

$$\Phi(E,R)=\frac{2\pi|\psi_m(R)|^2}{E_m-E}-\frac{1}{n}+2\int\limits_{2R/n}^{\infty}\frac{\tilde{W}(\xi)W(\xi)}{\xi^2}d\xi. \tag{27}$$

By means of the asymptotic series[9] of the functions $\tilde{W}$ and W, we obtain the expansion

$$\begin{aligned}-\frac{1}{n}+2\int\limits_{2R/n}^{\infty}\frac{\tilde{W}W}{\xi^2}d\xi&\cong-\frac{1}{n}+\frac{n}{R}+\frac{n^3}{2R^2}+\frac{n^5}{2R^3}+\frac{n^5+5n^7}{8R^4}+\ldots\\&=-\left(\frac{1}{n^2}-\frac{2}{R}\right)^{1/2}-\frac{\alpha_-}{R^4}+O(R^{-5});\quad\alpha_-=\frac{n^4}{4},\end{aligned} \tag{28}$$

which with an accuracy to R^{-5} is the sum of the expansion of the radical written out above and the polarization interaction of the negative and positive ions. The expressions for the negative ion polarizability α_- agrees with the expression obtained in ref. 12. Substituting Eq. (28) into (27), we obtain Eq. (7). The polarizability of the atomic core can be neglected, since it is much less than the polarizability of the negative ion.

In conclusion the authors express their gratitude to O. B. Firsov and M. A. Leontovich for an extremely helpful discussion of questions involved in this work.

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Translated by C. S. Robinson.
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