TUNNEL RECOMBINATION OF PROTONS NEAR THE SURFACE OF A METAL

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The tunnel recombination of protons near the surface of a metal is considered. Recombination probabilities to the ground level and the n-th level of a hydrogen ion are calculated for $v \ll v_F$ and $v \gg v_F$, where v is the velocity of the proton and v_F the velocity of the electron on the Fermi surface of the metal. The results are compared with the experiments of Phillips.[2]

1. INTRODUCTION

In a number of experiments,[1,2] the recombinations of ions coming up to a metal or passing through a thin metallic foil have been studied. The recombination process can take place in two ways here—by a tunnel transition of the electron from the metal to a level with the same energy in the ion (isoenergetic transition) and by transition to a lower level with transfer of the excess energy to the second electron, which can leave the metal (Auger effect). In the present paper, only transitions of the first type are considered; these are not accompanied by secondary electron emission.

The theory of such tunnel recombination was developed in the papers of Oliphant and Moon,[1] Massey,[3] Shekhter,[4] Cobas and Lamb,[5] and a number of others. In all the works mentioned, the calculation was carried out in two steps. First, it was assumed that the ion is at rest at a distance R from the surface of the metal (Fig. 1), and the probability of transition of the electron from the metal to the ion per unit time, $\nu(R)$, was calculated by time-independent perturbation theory. The total recombination probability is $w = \int \nu\, dt$, where $dt = dR/v$ (v is the speed of the ion). Thus, the first step is inconsistent, since the motion of the ion is not taken into account at all. This approach will be called what follows the "fixed ion approximation" (FIA).

In the present research, this inconsistency is removed by the use of time-dependent perturbation theory, which permits us to determine the recombination probability w for an arbitrary speed of the ion (Sec. 3). In particular, it is shown that the fixed ion approach leads to the correct result only for $v \ll v_F$, where v_F is the speed of the electrons on the Fermi surface, while for $v \gtrsim v_F$, the recombination probability can be materially different from that obtained within the FIA framework. For example, for $v \gg v_F$, there is an asymptotic dependence $w \sim 1/v^8$ (Sec. 4). We also present here a correction of the calculation of Shekhter[4] (see Sec. 2), who determined w within the framework of FIA, but made a mistake which increases the value of w by about three orders of magnitude.

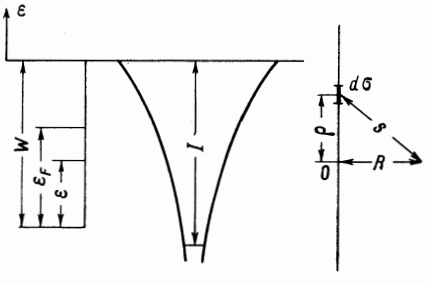

FIG. 1. Ion close to a metallic surface.

2. FIXED ION APPROXIMATION

Qualitatively, the probability of tunnel recombination can be estimated from the following semiclassical considerations (see Fig. 1). A number of electrons

$$dv = d\sigma \cos\theta \cdot 4m\varepsilon\, d\varepsilon\, d\Omega/(2\pi\hbar)^3$$

is incident on an area $d\sigma$ from the depth of the metal to its surface. Multiplying this quantity by the transmission coefficient of the barrier

$$D \approx \exp\left[-\frac{2}{\hbar}\int_{r_{min}} dr \sqrt{2m(|W|-\varepsilon-e^2/r)}\right]$$

$$\approx \exp\left[-\frac{2s}{\hbar}\sqrt{2m(|W|-\varepsilon)}\right], \qquad (2.1)$$

and integrating over the energy and the solid angle, we get

$$\nu(R) = \gamma\frac{\varepsilon_F}{\hbar}\exp\left(\frac{2R}{\hbar}\sqrt{2m\varphi}\right). \qquad (2.2)$$

Here, it is taken into account that

$$r_{min} = e^2/(|W| - \varepsilon),\ \cos\theta = R/s,\ d\sigma = 2\pi\rho d\rho,$$

$$s = \sqrt{\rho^2 + R^2}.$$

In (2.2), the quantity $\varphi = |W| - \epsilon_F$ is the external work function and γ is an undetermined quantity of order unity, which takes into account the inaccuracy in the determination of D and integration over $d\Omega$. The change in the number of ions is described by the equation

$$dN_i = -N_i \nu dt,\quad N_i = N_i(0)(1-w), \tag{2.3}$$

whence we find for the recombination probability

$$w = 1 - \exp\left(-\int_0^\infty \nu\, dt\right) = 1 - \exp(-\text{const}/v), \tag{2.4}$$

where const $= {}^1/_2 \gamma v_F \sqrt{\epsilon_F/\varphi}$. Thus, the w(v) dependence always has the standard form (2.4) in the FIA. In what follows, we shall show that a systematic account of the motion of the ion leads to another w(v) dependence for $v \gtrsim v_F$.

Let us now consider the quantum-mechanical formulation of the problem. Following Shekhter,[4] we have for the transition probability of a single electron per unit time

$$\nu = \frac{2\pi}{\hbar}|V_n|^2 \delta(E^0 - E^k),\qquad V_n = \int d\mathbf{r}\,\psi^*(\mathbf{r})\frac{e^2}{r}\chi(\mathbf{r}), \tag{2.5}$$

where V_n is the matrix element of the perturbation operator e^2/r for the transition of the electron from the metal to the n-th level of the ion. The wave functions of the electron in the hydrogen atom have the usual form (see, for example, [6]), and the wave function in the metal is equal to (see [4])

$$\chi(\mathbf{r}) = \begin{cases} A(e^{ik_x x} + a_k e^{-ik_x x})e^{i(k_y y + k_z z)}, & x < 0 \\ A b_k e^{-\delta x} e^{i(k_y y + k_z z)}, & x > 0 \end{cases}, \tag{2.6}$$

where $\delta = \sqrt{k_0^2 - k_x^2}$, $k_0^2 = 2m|W|/\hbar^2$, $A^2 = N$ is the density of electrons in the metal, $a_k = (ik_x + \delta)/(ik_x - \delta)$ is the amplitude of the reflected wave, while the amplitude b_k is determined from the matching conditions and is equal to $b_k = 2ik_x/(ik_x - \delta)$.

Let us consider the transition of the electron from the metal to the ground state of the hydrogen atom. The matrix element in this case was computed by Shekhter:

$$V_1 = \frac{2\sqrt{\pi}Ab_k e^2}{\sqrt{a_0^3}}\left\{\frac{e^{-\xi R}}{\xi(\delta - \xi)} - \frac{2e^{-\delta R}}{\delta^2 - \xi^2} + \frac{\delta + \xi}{k^2 + 1/a_0^2}\frac{e^{-\xi R}}{\xi}\right\}. \tag{2.7}$$

Here we have introduced the following notation:

$$\xi = \sqrt{1/a_0^2 + k_\perp^2},\quad k_\perp^2 = k_y^2 + k_z^2,\quad k^2 = 2m\varepsilon/\hbar^2.$$

The function $\delta(E^0 - E^k)$ appearing in Eq. (2.5) expresses the law of energy conservation and will be represented (in our notation) in the form

$$\delta(E^0 - E^k) = \delta(|W| - I - \varepsilon) = \frac{2m}{\xi}\delta(\xi - \delta). \tag{2.8}$$

Setting $\xi = \delta$ in (2.7), we find

$$V_1 = \frac{2\sqrt{\pi}Ab_k e^2}{\sqrt{a_0^3}} e^{-\xi R}\left(\frac{1}{2\xi^2} + \frac{2}{k_0^2}\right) \tag{2.9}$$

(as will be shown in Sec. 3, the dependence on R is taken into consideration only in the exponent).

Substituting (2.9) in (2.5) and averaging over the Fermi distribution, we find the probability of transition per unit time:

$$\langle \nu(R)\rangle = \left(\int_0^{\varepsilon_F} d\varepsilon\sqrt{\varepsilon}\right)^{-1}\frac{2\pi}{\hbar}\int_0^{\varepsilon_F} d\varepsilon\sqrt{\varepsilon}|V_1|^2\delta[\varepsilon - (|W| - I)]. \tag{2.10}$$

In particular, if the quantity $\epsilon = |W| - 1$ lies outside of the interval $(0; \epsilon_F)$, then the transitions are impossible. For the calculation of the total probability, it is necessary to integrate $\langle\nu(R)\rangle$ over $dt = dR/v$.

By using the value of $|W| = 18$ eV, assumed in [4] for a molybdenum surface for a proton speed $v = 1.5 \times 10^6$ cm/sec, we find $\int_0^\infty dt\,\langle\nu(R)\rangle \approx 10^2$, which is three orders smaller than the result of Shekhter.[1)]

According to recent data, $|W| \approx 10$ eV for molybdenum; moreover, one can assume for all metals that $|W| < 13.5$ eV. This means that, in averaging over the Fermi distribution (2.10), the zero point $\delta(\epsilon - |W| + I)$ lies outside the integration interval, and $w_1 = 0$ in the FIA. Therefore, we have computed the probabilities w_2 and w_3 for the electron transition to the second and third levels in the hydrogen atom, which, with account of (2.3) and (2.4), are given by

$$w_2 = 1 - \exp(-0.2e^2/\hbar v),\quad w_3 = 1 - \exp(0.08e^2/\hbar v). \tag{2.11}$$

The numerical factors 0.2 and 0.08 were obtained as the result of substitution of the value $|W| = 10$ eV and $\epsilon_F = 6$ eV, which are typical for many metals. These parameters are shown in the table, the data in which were taken from various sources.[7-9]

It is seen from the table that $\varphi = |W| - \epsilon_F$ exceeds the ionization energy of the fourth level I_4

[1)] The discrepancy is due to an arithmetic error made in [4].

	Cu	Ag	Au	Ca	W	Mo	Mg
$\|W\|$, eV	11.1	10.2	10.3	7.0	11.0	10.7	10.5
ε_F, eV	7.0	5.5	5.5	4.3	6.5	6.5	7.1
φ, eV	4.1	4.7	4.8	2.7	4.5	4.2	3.4

≈ 0.8 eV, and therefore the consideration of the transitions to the fourth and higher levels is not of interest.

3. ACCOUNT OF THE MOTION OF THE ION

The transition probability per unit time ν was calculated above under the assumption that the proton is found at a distance R from the boundary of the metal and can be considered to be at rest. This is why we used Eq. (2.5) from the theory of transitions for stationary excitation.

However, if the velocity of the ion is sufficiently large, and in particular, if it exceeds the velocity of the electron on the Fermi surface ($v \gtrsim v_F$), then it is obviously no longer possible to consider the excitation as stationary, and Eq. (2.5) is not valid. In this case, the transition probability must be computed from the Schrödinger equation, which takes into account the change of state with time.

$$\hat{H}\Psi = i\hbar\frac{\partial\Psi}{\partial t}; \quad \hat{H} = -\frac{\hbar^2}{2m}\Delta + U(\mathbf{r}, t). \tag{3.1}$$

Here

$$U(\mathbf{r}, t) = U_{\text{met}}(\mathbf{r}) + U_{\text{coul}}(\mathbf{r}, t) = \begin{cases} -e^2/|\mathbf{r} - \mathbf{v}t|, & x > 0 \\ -|W|, & x < 0. \end{cases} \tag{3.2}$$

The wave function of the electron can be represented in the form

$$\Psi(\mathbf{r}, t) = \alpha(t)\chi(\mathbf{r}, t) + \beta(t)\psi(\mathbf{r}, t). \tag{3.3}$$

Here

$$\chi(\mathbf{r}, t) = \chi(\mathbf{r})\exp\left[-\frac{i}{\hbar}(-|W| + \varepsilon)t\right],$$

$\chi(\mathbf{r})$ is determined by Eq. (2.6), and ψ is the hydrogen wave function. In the laboratory system of coordinates, the function $\psi(\mathbf{r}, t)$ is equal to (see [10])

$$\psi(\mathbf{r}, t) = \frac{1}{\sqrt{\pi a_0^3}}\exp\left\{-\frac{|\mathbf{r} - \mathbf{v}t|}{a_0} + i\frac{I}{\hbar}t + i\left(\frac{mv}{\hbar}x - \frac{mv^2}{2\hbar}t\right)\right\}. \tag{3.4}$$

The coefficients $\alpha(t)$ and $\beta(t)$ in (3.3) determine the probability of the state of the electron at a given instant of time in the metal or in the moving hydrogen atom.

Let us consider the case corresponding to the experiment of Phillips,[2] in which it can be assumed that at the initial instant of time (t = 0) the proton leaves the metal and goes off to infinity with the velocity v, which is equivalent to the conditions $\alpha(0) = 1$, $\beta(0) = 0$.

Assuming the Coulomb excitation $-e^2/|\mathbf{r} - \mathbf{v}t|$ to be small, we find from the Schrödinger equation

$$\dot{\beta}(t) = \frac{1}{i\hbar}\int d\mathbf{r}\,\psi^*(\mathbf{r}, t)\left(-\frac{e^2}{|\mathbf{r} - \mathbf{v}t|}\right)\chi(\mathbf{r}, t). \tag{3.5}$$

Carrying out the integration, we represent $\beta(t)$ in the form

$$\dot{\beta}(t) = B\exp\left\{-i\omega t + i\frac{k_p R}{2} - \xi R\right\}[f_v + g_v\exp\{-ik_p R + R(\xi - \delta)\}]. \tag{3.6}$$

Here, we have introduced the notation

$$k_p = \frac{mv}{\hbar}, \quad R = vt, \quad B = \frac{2\pi A b_k e^2}{\hbar\sqrt{\pi a_0^3}},$$

$$\omega = \frac{1}{\hbar}(I - |W| + \varepsilon),$$

$$f_v = \frac{1}{\xi}\left[\frac{1}{k_p + i(\xi - \delta)} - \frac{k_p + i(\xi + \delta)}{(k_p + i\xi)^2 + k_x^2}\right],$$

$$g_v = \frac{2i}{(k_p - i\delta)^2 + \xi^2}.$$

To determine the recombination probability in first-order perturbation theory, we must find

$$\beta(\infty) = \int_0^\infty dt\,\dot{\beta}(t), \quad w = \langle|\beta(\infty)|^2\rangle. \tag{3.7}$$

It is curious to see how Shekhter's result can be obtained from (3.6). This result (corrected by us) corresponds to the fixed-ion approximation. For this purpose, we must set $k_p = 0$ and R = const in (3.6). Then, as is not difficult to prove, (3.6) takes the form

$$\dot{\beta} = \frac{i}{\hbar}V(R)e^{-i\omega t},$$

where V(R) is identical with the matrix element of Shekhter (2.7). By computing

$$\beta(t) = \int_0^t dt\,\dot{\beta} = V(R)(1 - e^{-i\omega t})/\hbar\omega,$$

we get for the transition probability per unit time

$$\nu(R) = \lim_{t\to\infty}\frac{1}{t}|\beta(t)|^2 = \left|\frac{1}{\hbar}V(R)\right|^2 2\pi\delta(\omega), \tag{3.8}$$

which is identical with (2.5). Furthermore, by averaging (3.8) over the Fermi distribution and integrating over dt = dR/v, we get Eq. (2.11). In first-order perturbation theory, we can write this

in the form

$$w = \int \langle \nu(R) \rangle \, dt = \text{const}/v, \quad \text{const} = \int \langle \nu(R) \rangle \, dR. \quad (3.9)$$

Thus, in the fixed-ion approximation, we assume in the integral $\int dt\, \beta$ that R = const and v = 0, and for the next integration $\int \langle \nu(R) \rangle \, dt$, we assume R = vt and $v \neq 0$, which is inconsistent and which can lead to an error in a number of cases.

Thus, for example, in the indicated approximation, transitions to a level whose energy lies outside the interval $(-|W|; -|W| + \epsilon_F)$ are shown to be impossible, while in the theory which takes into account the motion of the ion, the energy conservation law does not hold (the perturbation is time-dependent), and also the transitions can exist. In particular, for a transition to the ground state, we find from (3.6) and (3.7):

$$\beta(\infty) = \int_0^\infty \dot{\beta}(t)\, dt = \frac{B}{\xi v + i(\omega - {}^1/_2 k_p v)} (f_v + g_v h_v^{(\omega)}),$$

$$h_v^{(\omega)} = \frac{\xi v + i(\omega - {}^1/_2 k_p v)}{\delta v + i(\omega + {}^1/_2 k_p v)} \quad (3.10)$$

and finally,

$$w^{(v)} = \left\langle \frac{B^2}{(\xi v)^2 + (\omega - {}^1/_2 k_p v)^2} |f_v + g_v h_v^{(\omega)}|^2 \right\rangle. \quad (3.11)$$

In view of the complicated character of this expression, we consider two limiting cases, $v \gg v_F$ and $v \ll v_F$, for which we can find

$$f_v + g_v h_v^{(\omega)} = \begin{cases} f_0 + g_0 \approx 2i/k_0^2, & v \ll v_F \\ -2i/k_p^2 & v \gg v_F \end{cases}. \quad (3.12)$$

The expression (3.11) can be represented in the form

$$w_1^{(v)} = \frac{4^4}{3\pi} \left(\frac{e^2}{\hbar v_F}\right)^5 \left(\frac{\varepsilon_F}{|W|}\right)^3 F\left(\frac{v}{v_F}\right),$$

$$F\left(\frac{v}{v_F}\right) = \left\langle \frac{k_x^2}{k_F^2} \frac{k_0^4}{4} \frac{|f_v + g_v h_v^{(\omega)}|^2}{q^2 + (\varepsilon/\varepsilon_F - z_p)^2} \right\rangle. \quad (3.13)$$

We have here introduced the following notation:

$$q = \frac{\hbar \xi v}{\varepsilon_F} \approx \frac{\hbar \bar{\xi} v}{\varepsilon_F},$$

$$z_p = \frac{|W| - I + mv^2/2}{\varepsilon_F}.$$

For small velocities, and with account of (3.12), the function $F(v/v_F)$ takes the form

$$F\left(\frac{v}{v_F}\right) = \frac{1}{2} \int_0^1 \frac{dz\, z^{3/2}}{q^2 + (z - z_p)^2} \quad (3.14)$$

and remains finite even as $v \to 0$. Figure 2 shows a plot of $F(v/v_F)$ for typical values of the parameters: I = 13.5 eV, |W| = 10 eV, ϵ_F = 6 eV, and

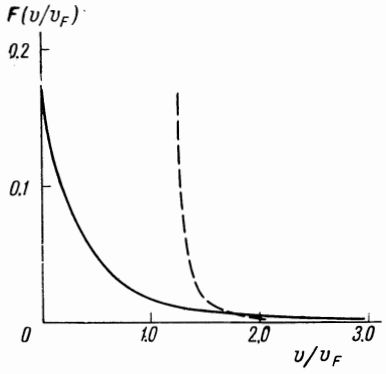

FIG. 2. Dependence of the recombination probability on the velocity of the ion. The dashed curve corresponds to the asymptotic formula (3.15).

$\bar{\xi} = 1.25/a_0$. Here the value of w_1 is seen to be ~10 for v = 0, which indicates the inadequacy of the first-order perturbation theory for velocities close to zero (according to the meaning of probability, w cannot be larger than unity). Therefore, we can use Eq. (3.13) only for $w \ll 1$.

In Phillips' experiments,[2] the passage of protons with energy in the range E_p = 5–200 keV through a thin metallic foil was studied. The corresponding value of v was $(0.7–4.5)v_F$. It is clear that under these conditions the fixed-ion approximation can lead to large errors. In the opposite limiting case, $v \gg v_F$, we find (with the help of (3.12))

$$F\left(\frac{v}{v_F}\right) \cong \frac{1}{5} \left(\frac{|W|}{\varepsilon_F}\right)^2 \left(\frac{v_F}{v}\right)^8 \approx 0.6 \left(\frac{v_F}{v}\right)^8. \quad (3.15)$$

This curve is shown as the dashed line in Fig. 2. The asymptote of (3.15) gives correct values only beginning with $v/v_F \gtrsim 4$; therefore comparison with experiment is difficult.

Let us compare the resultant $w_1^{(V)}$ dependence with the data of Phillips. Figure 3 (solid curve) describes the experimental dependence of the number of unrecombined protons on their energy E_p. The dashed curve $(1 - w_1^{(V)})$ corresponds to the approximate formula (3.14). Comparison shows that the recombination probability $w_1(v)$ (3.14) gives a somewhat lower value than experiment. Evidently this is explained by the fact that there exist other mechanisms responsible for the change in the charge ratio of the particles coming from the foil, for example, Auger recombination, connected with the emission of a secondary electron, and also triple recombination.[11]

4. TUNNEL TRANSITION OF AN ELECTRON TO THE n-TH LEVEL OF A HYDROGEN ATOM

Up to now we have considered the tunnel transition of an electron of a metal to the first level of a

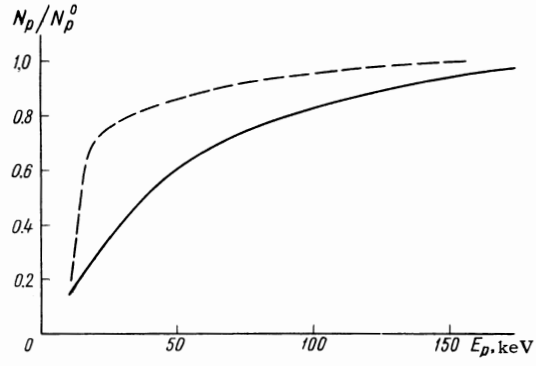

FIG. 3. Comparison of the theoretical data (dashed curve) with the experiments of Phillips.[2]

proton moving with velocity v. However, there is a finite probability of a tunnel transition to the second, third, etc. levels, with formation of an excited hydrogen atom. Limiting ourselves for simplicity only to spherically symmetric states ($l = 0$), we write down the eigenfunction of the n-th level in the form (see [10,12])

$$\psi_n^*(\mathbf{r},t) = -\frac{\exp[it\hbar^{-1}(I/n^2 - mv^2/2) + ik_p x]}{2\sqrt{\pi a_0}\, n!\sqrt{n}} \times \left\{\left(\frac{d}{d\gamma}\right)^n \gamma^{n-1}\left[\frac{\exp\{-|\mathbf{r}-\mathbf{v}t|(2\gamma-1)/na_0\}}{|\mathbf{r}-\mathbf{v}t|}\right]\right\}, \quad (4.1)$$

where γ is a parameter which, after differentiation, must be set equal to unity. Substituting (4.1) in (3.5) and integrating with respect to time, we get

$$\beta_n(\infty) = -\frac{2\sqrt{\pi}\, e^2 mAb_k}{\hbar^2\sqrt{a_0}\, n!\sqrt{n}}\left(\frac{d}{d\gamma}\right)^n \gamma^{n-1} \int\limits_{\Phi_n=(2\gamma-1)/na_0}^{\infty} d\Phi \times \{[\sqrt{\Phi^2+k_\perp^2}(k_p - i\delta + i\sqrt{\Phi^2+k_\perp^2})(2k_p\sqrt{\Phi^2+k_\perp^2} + i/(na_0)^2 - ik_p^2 - ik_0^2 + ik^2]^{-1} + 2i[[(k_p - i\delta)^2 + k_\perp^2 + \Phi^2](2k_p\delta + i/(na_0)^2 + ik_p^2 - ik_0^2 + ik^2)]^{-1} - (k_p + i\delta + i\sqrt{\Phi^2+k_\perp^2})[\sqrt{\Phi^2+k_\perp^2} \times((k_p + i\sqrt{\Phi^2+k_\perp^2})^2 - k_x^2)(2k_p\sqrt{\Phi^2+k_\perp^2} + i/(na_0)^2 - ik_p^2 - ik_0^2 + ik^2)]^{-1}\}. \quad (4.2)$$

In view of the complicated character of this expression, we limit ourselves to the highest velocities of the proton, $v \gg v_F$. Then (4.2) takes the form

$$\beta_n(\infty) = \frac{2i\sqrt{\pi}\, e^2 mAb_k}{\hbar^2\sqrt{a_0}\, k_p^3 n!\sqrt{n}}\left(\frac{d}{d\gamma}\right)^n \gamma^{n-1}\ln\frac{\Phi_n - ik_p}{\Phi_n + ik_p}. \quad (4.3)$$

In particular,

$$M_-(n) = \left(\frac{d}{d\gamma}\right)^n \gamma^{n-1}\ln(\Phi_n - ik_p) = -(n-1)!\left[\left(1 - \frac{2}{1 - ink_p a_0}\right)^n - 1\right]. \quad (4.4)$$

In the limit of high velocities ($k_p a_0 \gg 1$), one gets, approximately,

$$M_-(n) \approx (n-1)!\frac{2i}{k_p a_0}. \quad (4.5)$$

Similarly,

$$M_+(n) = \left(\frac{d}{d\gamma}\right)^n \gamma^{n-1}\ln(\Phi_n + ik_p) \approx -(n-1)!\frac{2i}{k_p a_0} \quad (4.6)$$

and then

$$\beta_n(\infty) = -\frac{8\sqrt{\pi}\, e^2 mAb_k}{\hbar^2 a_0^{3/2} k_p^4 n^{3/2}}. \quad (4.7)$$

Computing $|\beta_n(\infty)|^2$ and averaging over the Fermi distribution, we get

$$w_n^{(v)} = w_1^{(v)}/n^3, \quad (4.8)$$

where $w_1^{(v)}$ is determined by Eq. (3.13), while one must use the asymptote (3.15) for $F(v/v_F)$. We note that the $1/n^3$ dependence holds also for the radiative recombination of the electron in a gas.[13]

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