Proton neutralization at the surface of a solid

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Making use of the Bates-type quantum-mechanical recombination theory, the probability of the proton neutralization at the surface of a solid is discussed. The neutral fraction for the transition to the ground state of the hydrogen atom is calculated, and the dependences of the neutral fraction on the exit energy and angle of proton emerging from a metal are discussed. Numerical calculations are performed for Au, and the results are compared with experiments.

I. INTRODUCTION

During the past ten years, many workers have performed experiments to investigate charge states in ion beams passing through solids.[1,2] Recently, with increasing effort, experiments with light-ion beams, especially protons or deuterons, have been performed for various target materials by use of transmitting,[3,4] backscattering[5] and surface scattering[6] techniques, and several experimental results have been reported.

(i) The observed neutral fractions (or charged fractions) of proton and deuteron beams are weakly dependent on target materials.[3,5,7]

(ii) In the backscattering experiments for proton beams, the neutral fractions measured for a fixed energy and angle of emergence, are almost independent of the energy and angle of incidence within experimental errors.[5]

(iii) In the transmitting experiment with helium-ion beams, the observed result for incidence parallel to the channeling direction is almost identical to the result for incidence in random direction.[4]

(iv) In experiments on polarized-electron capture by a deuteron, a high polarization of the deuterium atoms has been observed under the channeling condition,[8] and has also been obtained under the surface scattering condition.[6]

Generally speaking, from these experimental results, it is pointed out the weak dependence of charge states in light-ion beams on experimental methods, or conditions. This suggests that electron capture by light ions, especially protons or deuterons, mainly takes place near the solid's surface,[7,9] and is of considerable interest in the study of electronic states in the surface region. However, the above suggestion has not been completely explained theoretically.

Theoretical studies in this field have been devoted to discuss the existence of a stable bound-electronic state in the ion inside metals, rather than the probability of electron capture or loss. Rogers *et al.*[10] and Ebel[7] have discussed stable bound-electronic states in the ion, taking into account the screened field created by many-body effects inside the metal. They concluded that, for the case of the hydrogen atom, there can not exist stable bound states inside the metal in both the static and dynamical cases. This is a negative conclusion concerning the possibility of electron capture inside metals.

As far as the theoretical treatment of the probability of electron capture or loss by protons passing through the solid is concerned, there is a major difficulty arising from the fact that the charge-transfer process is a three-body problem. Due to this difficulty, theoretical studies of this type are still statistical and phenomenological.[9,11]

In this paper, on the basis of Bates-type quantum-mechanical recombination theory,[12,13] we discuss the probability of the proton neutralization at the solid's surface.

II. DERIVATION OF THE RECOMBINATION PROBABILITY

We consider a collision in which the electron is initially in a metal, and finally is bound in the nth excited state of energy E_n to a hydrogen atom. Taking into account the experimental suggestion and the theoretical studies performed by Rogers *et al.* and Ebel mentioned in Sec. I, we assume that the transition takes place at the surface where the screening effect diminishes enough for bound states to be stable.[9] In describing the electron in the metal, we use a planar uniform-background model,[14] and take the depth of the square-well potential at the surface as U_0. Then, taking into account the motion of the proton, we start from the time-dependent Schrödinger equation. Following

the method formulated by Bates[12,13] and using the two-state approximation, we write down

$$ih\frac{\partial\Phi}{\partial t}=H\Phi\,, \qquad (1)$$

$$H=-(\hbar^2/2m)\Delta+V(\vec{r},t)+(-U_0)\theta(-z)\,,$$

$$\Phi(\vec{r},t)=C_1(t)\phi_1(\vec{r},t)+C_2(t)\phi_2(\vec{r},t)\,, \qquad (2)$$

where $V(\vec{r},t)=-e^2/|\vec{r}-\vec{v}_p t|$, and θ the Heaviside step function. We take the direction normal to the surface as z ($z<0$, inside the metal; $z>0$, outside the metal). At the time $t=0$, the proton reaches the metal surface. In the above, m and $\vec{r}$ are the mass and the coordinate of the electron, respectively, and $\vec{v}_p$ the velocity of the proton which is taken to be constant as far as the determination of the recombination probability is concerned. The C's are coefficients dependent on the time t and $\phi_1(\vec{r},t)$ and $\phi_2(\vec{r},t)$ the wave functions of the electron in the metal and in the hydrogen atom, respectively, which are given by,

$$\phi_1(\vec{r},t)=\phi_0(\vec{r})e^{-(i/\hbar)(-U_0+\hbar^2k^2/2m)t}\,, \qquad (3)$$

$$\phi_0(\vec{r})=\begin{cases}A(e^{ik_z z}+Be^{-ik_z z})e^{i(k_x x+k_y y)}\,, & z<0\\ ACe^{-\delta z}e^{i(k_x x+k_y y)}\,, & z>0\end{cases} \qquad (4)$$

and

$$\phi_2(\vec{r},t)=\phi_n(|\vec{r}-\vec{v}_p t|)e^{(i/\hbar)(m\vec{v}_p\cdot\vec{r}-E_n t-mv_p^2 t/2)}\,, \qquad (5)$$

with

$$\delta=(k_0^2-k_z^2)^{1/2},\quad k_0^2=2m\hbar^{-2}U_0\,,\quad A^2=2/\Omega$$

$$B=\frac{ik_z+\delta}{ik_z-\delta},\quad C=\frac{2ik_z}{ik_z-\delta}\,, \qquad (6)$$

Ω being the volume of our macroblock, ϕ_n the nth excited-state wave function of the hydrogen atom.

Substituting Eq. (2) into the Schrödinger equation, we obtain the coupled differential equation for C_1' and C_2',

$$i\hbar\dot{C}_1'(t)=\epsilon_{12}(t)C_2'(t)\exp\left(-\frac{i}{\hbar}\int^t[\epsilon_2(t)-\epsilon_1(t)]dt\right) \qquad (7)$$

and

$$i\hbar\dot{C}_2'(t)=\epsilon_{21}(t)C_1'(t)\exp\left(-\frac{i}{\hbar}\int^t[\epsilon_1(t)-\epsilon_2(t)]dt\right)\,, \qquad (8)$$

where

$$C_1(t)=C_1'(t)\exp\left(-\frac{i}{\hbar}\int^t\frac{h_{11}-S_{12}h_{21}}{1-|S|^2}dt\right)\,, \qquad (9)$$

$$C_2(t)=C_2'(t)\exp\left(-\frac{i}{\hbar}\int^t\frac{h_{22}-S_{21}h_{12}}{1-|S|^2}dt\right), \qquad (10)$$

$$\epsilon_{12}(t)=\frac{1}{1-|S|^2}(h_{12}-S_{12}h_{22})=\epsilon_{12}'(t)e^{-(i/\hbar)(E_2-E_1)t}\,, \qquad (11)$$

$$\epsilon_{21}(t)=\frac{1}{1-|S|^2}(h_{21}-S_{21}h_{11})=\epsilon_{21}'(t)e^{-(i/\hbar)(E_1-E_2)t}\,, \qquad (12)$$

and

$$\epsilon_2(t)-\epsilon_1(t)=\frac{1}{1-|S|^2}(h_{22}-h_{11}-S_{21}h_{12}+S_{12}h_{21})\,, \qquad (13)$$

with

$$S_{12}=\langle\phi_1|\phi_2\rangle=S_{21}^*,\quad |S|^2=S_{12}S_{21}\,,$$

$$h_{12}=\langle\phi_1|(-U_0)\theta(-z)|\phi_2\rangle,\quad h_{21}=\langle\phi_2|V|\phi_1\rangle\,,$$

$$h_{11}=\langle\phi_1|V|\phi_1\rangle\,,\quad h_{22}=\langle\phi_2|(-U_0)\theta(-z)|\phi_2\rangle\,, \qquad (14)$$

$$E_1=-U_0+(\hbar^2k^2/2m)\,,\quad E_2=E_n+\tfrac{1}{2}mv_p^2\,,$$

where the dot means the derivative for t, and $\langle\cdots\rangle$ the integration over $\vec{r}$.

If S_{12}, S_{21}, h_{11}, and h_{22} are neglected, and $C_1'(t)$ are approximated to unity (the first-order Born approximation), Eq. (7) disappears, and Eq. (8) reduces to the result calculated by Trubnikov and Yablinskii.[15] Furthermore, if V is taken to be a static Coulomb potential (the fixed-ion model), Eq. (8) reduces to the result calculated by Massey[16] and others[17,18] using the time-independent theory.

From the coupled equations (7) and (8), we obtain

$$\ddot{C}_2'+\left(\frac{i}{\hbar}[\epsilon_1(t)+E_1-\epsilon_2(t)-E_2]-\frac{\dot{\epsilon}_{21}'(t)}{\epsilon_{21}'(t)}\right)\dot{C}_2'+\frac{\epsilon_{12}(t)\epsilon_{21}(t)}{\hbar^2}C_2'=0\,. \qquad (15)$$

In the following, we refer to the approximate method within the framework of the Bates-type method used here. As far as the post and prior interactions are concerned, we note that

$$\epsilon_{12}(t)\epsilon_{21}(t)=|(h_{12}-S_{12}h_{22})(h_{21}-S_{21}h_{11})|\,, \qquad (16)$$

provided that the equivalence of the post and prior interactions[12] is realized. The realization of the equivalence of the post and prior interactions is equivalent to the case of $\Delta E=0$ when $\epsilon_{12}\epsilon_{21}$ is expanded by ΔE ($\Delta E=E_1+h_{11}-E_2-h_{22}$). The above term $\Delta E=0$ represents a major contribution to the transition. In this paper, we avoid discussing the detailed problem relating to the post-prior discrepancy[12,19] because it is considerably complex and it is the challenging problem in the field in treating a collision in which electron exchange, or the charge-transfer processes are involved, and we neglect the contribution of the higher terms in ΔE to $\epsilon_{12}\epsilon_{21}$ as a first-order approximation, because

they are higher-order contributions to the transition which are considered to be small in comparison with the contribution from the major term $\Delta E = 0$.

In deriving the probability of the proton neutralization at the solid's surface, we made the following assumptions:

(i) The transition region is so small that during the transition $\epsilon_1 - \epsilon_2$ is regarded as a linear function of time, and $\epsilon'_{12}, \epsilon'_{21}$ (or $\epsilon_{12}, \epsilon_{21}$) as independent of time. Therefore, according to the experimental suggestion that the transition takes place at the solid's surface, we can expand $\epsilon_1(t) - \epsilon_2(t) \cong A_1 + A_2 t$ around $t = 0$, and replace $\epsilon_{21}(t)\epsilon_{12}(t)$ by $\epsilon_{21}(0)\epsilon_{12}(0)$, and $\dot{\epsilon}'_{21}$ by 0, where $A_1 = \epsilon_1(0) - \epsilon_2(0)$, and $A_2(d/dt)[\epsilon_1(t) - \epsilon_2(t)]_{t=0}$ [we note that A_2 is proportional to v_{pz} in general, where v_{pz} is the z component of the velocity of the proton (see also Sec. III)].

(ii) The overlap integrals S_{12} and S_{21} are neglected as a first-order approximation, because $|S|^2$ is much smaller than unity. h_{11} represents the energy-level shift of the electron in the metal affected by the field created by the proton (h_{22} that of the electron in the hydrogen atom affected by the metal field). As far as h_{11} is concerned, we take into account the experimental suggestion, and the theoretical works performed by Rogers *et al.* and Ebel mentioned in Sec. I, and in the problem discussed here neglect the contribution to h_{11} from the electrons inside the metal because it is considered to be small due to the screening effect inside the metal.

Then, according to the above assumptions and Eq. (16), and substituting $t' = (E_1 - E_2 + A_1)/A_2 + t$, we obtain from Eq. (15)

$$\ddot{C}'_2 + (i/\hbar)A_2 t' \dot{C}'_2 + (\beta/\hbar^2)C'_2 = 0 , \tag{17}$$

where $\beta = |h_{12}h_{21}|$ at $t = 0$.

The boundary conditions appropriate to the proton neutralization at the solid's surface being investigated here are

$$|C'_1(-\infty)| = 1 , \quad C'_2(-\infty) = 0 . \tag{18}$$

According to the Landau-Zener formula,[20] from Eqs. (17) and (18), we obtain the recombination probability

$$\phi_0 = |C'_2(\infty)|^2 = 1 - e^{-w} , \tag{19}$$

where $w = 2\pi\beta/\hbar|A_2|$.

In the above, if β is independent on $\vec{v}_p$, w is proportional to v_{pz}^{-1}. This condition is realized at the region where the velocity of the proton is slow enough.

III. TRANSITION TO THE GROUND STATE OF A HYDROGEN ATOM

In this section, we consider the transition of an electron in the metal to the first energy level of a proton moving with velocity $\vec{v}_p$. The eigenfunction of the ground state of the hydrogen atom is written in the form

$$\phi_{1s}(|\vec{r} - \vec{v}_p t|) = [1/(\pi a_0^3)^{1/2}]e^{-|\vec{r}-\vec{v}_p t|/a_0} , \tag{20}$$

where a_0 is the Bohr radius. We take the exit angle between the beam direction and the direction normal to the surface as θ. Then, $\vec{v}_p = (0, v_{py}, v_{pz})$, $v_{py} = v_p \sin\theta$, $v_{pz} = v_p \cos\theta$ and $v_p = (v_{py}^2 + v_{pz}^2)^{1/2}$.

Using Eqs. (3)–(5), (14), and (20), we obtain

$$h_{12}(0) = -\frac{2\sqrt{\pi}}{\sqrt{a_0^5}} A U_0 \frac{1}{\alpha^2} \times \left[\frac{1}{\alpha + i(q_{pz} - k_z)} \left(\frac{1}{\alpha} + \frac{1}{\alpha + i(q_{pz} - k_z)} \right) + B^* \frac{1}{\alpha + i(q_{pz} + k_z)} \left(\frac{1}{\alpha} + \frac{1}{\alpha + i(q_{pz} + k_z)} \right) \right] , \tag{21}$$

$$h_{21}(0) = -\frac{2\sqrt{\pi}}{\sqrt{a_0^3}} A e^2 \frac{1}{\alpha} \times \left(\frac{1}{\alpha - i(q_{pz} - k_z)} + \frac{B}{\alpha - i(q_{pz} + k_z)} + \frac{C}{\alpha + \delta + i q_{pz}} \right) , \tag{22}$$

and

$$A_2 \cong -[(U_0/2a_0) + A^2 e^2 |C|^2 (\pi/\delta)] v_{pz} , \tag{23}$$

where $\alpha = (a_0^{-2} + q_\perp^2)^{1/2}$, $\hbar\vec{q}_p = m\vec{v}_p$, and $\vec{q}_\perp = (k_x, k_y - q_{py})$.

Then, from Eqs. (21)–(23), we can obtain w. At first, we consider the normal-emergence case ($\theta = 0$). The v_{pz} dependence of w is simplified for two types of limiting cases. For the high-energy case ($v_{pz} \gg v_F$; v_F the Fermi velocity), we can find

$$\beta \propto v_{pz}^{-3} . \tag{24}$$

It follows

$$w \propto E_p^{-2} , \tag{25}$$

where E_p is the kinetic energy of the proton.

Furthermore, if $w \ll 1$, one obtains

$$\phi_0 \approx w \propto E_p^{-2} . \tag{26}$$

On the other hand, for the low-energy case ($v_{pz} \ll v_F$), as was indicated in Sec. II, β is independent of v_{pz}, from which follows

$$w \propto E_p^{-1/2} . \tag{27}$$

Next, we consider the θ emergence case. As is obtained from Eqs. (3)–(5), (14), and (20), $h_{12}(0)$

and $h_{21}(0)$ are written in the form of an integral representation over $\vec{r}_\perp$, that is

$$h_{12}(0)=\int d\vec{r}_\perp(\cdots)e^{-i\vec{q}_\perp\cdot\vec{r}_\perp} \tag{28}$$

and

$$h_{21}(0)=\int d\vec{r}_\perp(\cdots)e^{i\vec{q}_\perp\cdot\vec{r}_\perp}\,, \tag{29}$$

The phase factor $e^{i\vec{q}_\perp\cdot\vec{r}_\perp}$ appearing in Eqs. (28) and (29) means that the electron experiences a momentum transfer of $\hbar\vec{q}_\perp$ at the transition. When $\vec{q}_\perp$ is large, it vibrates violently, and makes the integrate over $\vec{r}_\perp$ small. Physically, it corresponds to the fact that a large momentum transfer is hard to obtain. Then, it is understood that in the higher-energy region ($q_{py}\gg k_F$), the transition of the electron is restricted. On the other hand, in the low-energy region ($q_{py}\ll k_F$), the effect of this phase factor diminishes. Chateau-Thierry and Gladieux[3] pointed out experimentally that the neutral fraction varies with the angle θ, and decreases with increasing θ at 1.4-MeV exit energy of the proton emerging from Au and Al.

In the high-energy region ($v_{py}\gg v_F$), we find

$$\beta\propto 1/v_{py}^3v_p^3\,. \tag{30}$$

This leads to

$$w\propto 1/v_{pz}v_{py}^3v_p^3=1/v_p^7\sin^3\theta\cos\theta\,. \tag{31}$$

Following Eq. (31), we obtain the result that ϕ_0, or the neutral fraction, decreases with increasing θ up to 60°. Furthermore, if $w\ll 1$, we have

$$\phi_0\approx w\propto 1/v_p^7\sin^3\theta\cos\theta\,. \tag{32}$$

On the other hand, in the low-energy region ($v_p\ll v_F$), one obtains as for Eq. (27),

$$w\propto 1/v_{pz}=1/v_p\cos\theta\,. \tag{33}$$

In this case, we obtain the result that ϕ_0, or the neutral fraction, increases with increasing θ. As far as the v_{pz} dependence is concerned, this result coincides with the result obtained by Cobas and Lamb,[17] and Hagstrum.[18]

In order to determine the neutral fraction in the two-state approximation, we average ϕ_0 over the Fermi distribution and find

$$\langle\phi_0\rangle=\left(\int_0^{\epsilon_F}d\epsilon\sqrt{\epsilon}\right)^{-1}\int_0^{\epsilon_F}d\epsilon\sqrt{\epsilon}\,\phi_0\,, \tag{34}$$

where $\epsilon=\hbar^2k^2/2m$ and ϵ_F if the Fermi energy.

For the high-energy case, if $w\ll 1$, we obtain from Eqs. (26) and (32),

$$\langle\phi_0\rangle\propto\begin{cases}E_p^{-2} & \text{(normal-emergence case)}\\ \dfrac{1}{v_p^7\sin^3\theta\cos\theta} & \end{cases} \tag{35}$$

(θ-emergence case for $v_p\sin\theta\gg v_F$).

Numerical calculations are performed for Au. In Fig. 1, the dashed curve and the short- and long-dashed curve show the results for the high-energy approximation [Eq. (25)] and the low-energy approximation [Eq. (26)], respectively. The solid curve shows the result for the exact numerical calculations. Experimental data obtained from transmitting experiments[3] are also shown in Fig. 1. Comparison between our results and experiments

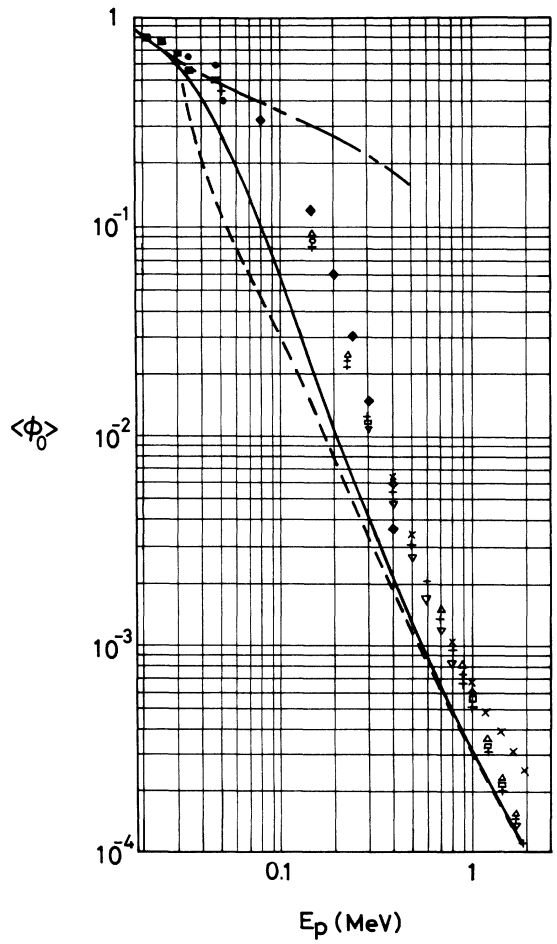

FIG. 1. Neutral fraction $\langle\phi_0\rangle$ for Au as a function of E_p in the case of normal emergence, where E_p is the exit energy of the protons. The dashed and dot-and-dashed curves are results of the high-energy approximation and the low-energy approximation, respectively. The solid curve is the result of exact numerical calculations. Experimental data obtained from transmitting experiments are also shown (Ref. 3).

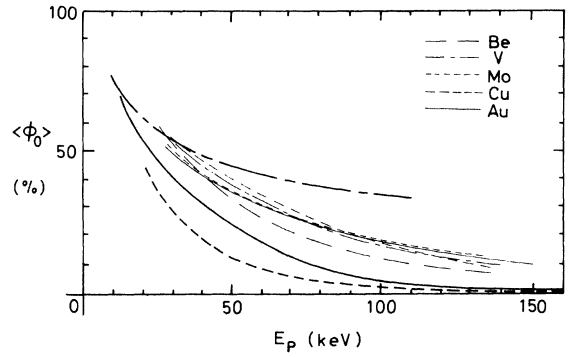

FIG. 2. Neutral fraction $\langle\phi_0\rangle$ for Au as a function of E_p in the case of normal emergence, where E_p is the exit energy of the protons. The dashed, dot-and-dashed, and solid curves represent same meanings as in Fig. 1. Experimental results for Be, V, Mo, Cu, and Au, which are obtained from backscattering experiments, are also shown (Refs. 5 and 21).

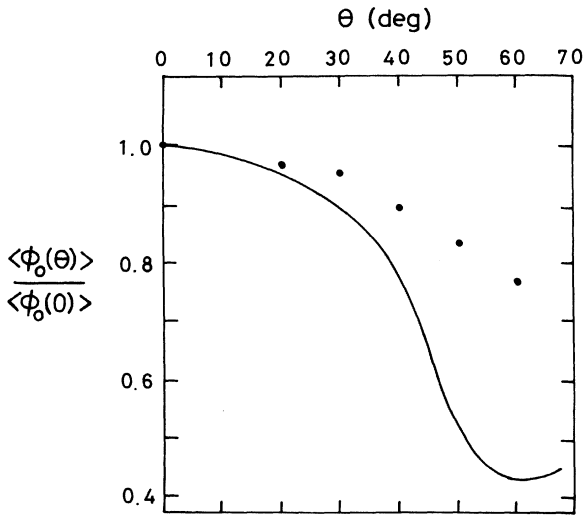

FIG. 3. Ratio $\langle\phi_0(\theta)\rangle / \langle\phi_0(0)\rangle$ as a function of the exit angle θ for Au at 1.4 MeV exit energy of the protons, represented by the solid curve. Experimental data are also shown (Ref. 3).

shows that our results give values of the right order of magnitude in a wide range of proton energy (10 keV–1.6 MeV). The energy dependence of our results is in good agreement with the experimental trend, especially in the high-energy region.

Figure 2 shows the results for proton energy up to about 150 keV. Experimental results for Be, V, Mo, Cu,[5] and Au,[21] which are obtained from backscattering experiments, are also shown. In the region where the proton energy is less than 10 keV, the low-energy approximation is good. In Figs. 1 and 2, our results give somewhat lower values than experiments.

Figures 1 and 2 show the normal emergence case. θ-emergence cases are also calculated. Figure 3 shows the dependence of $\langle\phi_0\rangle$ on θ for 1.4 MeV exit energy of the protons. Our results show that the neutral fraction decreases with increasing θ up to 60°. Qualitatively, this dependence is agreement with the experimental result.[3] The decrease we obtain is more rapid than the experimental one.

IV. CONCLUDING REMARKS

Making use of the Bates-type quantum-mechanical recombination theory, the proton neutralization at the solid's surface was studied, and the neutral fraction for the transition to the ground state of the hydrogen atom was calculated in detail. In deriving the transition probability, we introduced the Landau-Zener formula from a mathematical point of view.

Numerical calculations were performed for Au, and were compared with experiments. Calculated results gave values of right order of magnitude in a wide range of proton energies (10 keV–1.6 MeV), and their energy dependence was in good agreement with the experimental trend, especially in the high-energy region. We obtain the result that in the high-energy region, the neutral fraction is proportional to E_p^{-2}.

The θ-emergence cases were also calculated in the high-energy region by taking into account that a large momentum transfer experienced by the electron is hard to obtain when $v_{py} \gg v_F$. The calculated θ dependence of the neutral fraction is in agreement with the experiment performed by Chateau-Thierry and Gladieux up to 60°, qualitatively. In comparison with the experiment for 1.4 MeV exit energy of the proton emerging from Au, the θ dependence of our result was strong. The θ dependence in the high-energy region is contrary to that of Cobas and Lamb, and Hagstrum. The tendency of the neutral fraction to increase with increasing θ was realized in the low-energy region where our result coincides with that of Cobas and Lamb, and Hagstrum.

Finally, our results gave a somewhat lower value than experiments, which is naturally explained by the fact that there exist other processes relating to the proton neutralization at the solid's surface, i.e., transitions to the excited states of the hydrogen atom, Auger recombination, radiative recombination, etc. As far as the transitions to the excited states of the hydrogen atom are concerned, it is considered that they are smaller than the transition to the ground state as n^{-3},[15] where

n is the principal quantum number of the hydrogen atom. This n^{-3} dependence is well known in the case of radiative recombination[22] in a gas. Although the transitions to the excited states must be taken into account, they are considered to be higher-order corrections to the results calculated here. On the other hand, in order to explain the experiment completely, the contributions to the problem studied here from Auger recombination and radiative recombination must be taken into account.

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