Ps formation calculation scheme inside muffin-tins

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(Received 29 June 1990)

Abstructs :

Calculation scheme of positronium (Ps) formation inside muffin-tins is presented for calculation program code of angle-resolved Positronium foramtion spectroscopy (ARPsFS). Layered KKR method is used in this calculation Spectrum calculation of angle-resolved Ps formation will be performed for a lot of conditions by building in the new program code into the previous calculation where Ps formation is calculated only around surface barrier outside of the topmost muffin-tin. Subroutines for the present calculation are supplied in the angle-resolved photoemission spectrum calculation code, SLON established by Blake, Koukal and Larsson.

1. Introduction

Since positronium can be formed only outside of surface for metal and semiconductor, Ps is expected to be a good tool for surface spectroscopy [1]. Especially, angle-resolved Ps formation spectroscopy is expected to be a powerful spectroscopy to measure surface electronic structure, because its mechanism is quite similar to angle-resolved photoelectron spectroscoopy and ARPsFS is more sensitive to surface electronic structure [2-12].

In the last decade, technique for intense monoenergetic positron beam has been developed well [1,13]. Lawrence Livermore, Mainz University, Electronic Technology Laboratory (Denso-ken), Osaka University and JAERI Tokai have well-operated intense pulse positron beam lines. Using those machines, we can do very easily ARPsFS experiment. Technique to measure Ps energy distribution has been established for pulse beam [14,15].

However, to analyse experimental spectra, comparison with theory is necessary. Though we could expect that ARPsFS spectrum is similar to surface local density of states, we should encounter certain modification by so-called matrix element effect. Similar situations

are in ARUPS : we could analyse nothing without program code of ARUPS calculation [16]. Program code for ARPsFS has been developed by using theory of ref. 12. We included Ps formation only around the surface barrier (outside of the barrier and the space between the barrier and the topmost muffin-tin layer) in the theory presented in ref. 12. Though Ps wave function decays strongly towards inside from the barrier, it looks necessary to include at least terms from muffin-tins of the topmost atomic layer. It is also confirmed by results of ARUPS calculation, because we use similar calculation scheme for it.

Thus, the purpose of the paper is to build in terms inside the muffin-tins into the Ps formation theory. We will also explain how to build in the above teory into the previous program code with subroutines supplied in SLON.

2. Outline of the theory

Revolution to shorten computational time for ARUPS calculation was done by J. B. Pendry with his 'LEED-type calculation' [17]. Key point of hig theory is to use Green's function for initial electronic states : conversion procedure is saved by relaxation of hole remaining in the surface. In his formalism, photoelectron intensity,

$$I(\vec{k}_{\parallel}, E+w)=\sum_j \frac{L}{k_z^+} |<\vec{k}_{\parallel}, E+w|\triangle|j, E>|^2 \quad \cdots\cdots(1)$$

is rewritten into the following formula with the Green' function for initial states.

$$I(\vec{k}_{\parallel}, E+w)=-\frac{1}{\pi} Im\iint \psi_2(\vec{r})\triangle G_1^+(\vec{r}, \vec{r}', E)\triangle^+ \psi_2^*(\vec{r}')\, d^3\vec{r}\, d^3\vec{r}'$$

$$\triangle=-\frac{i\vec{a}\cdot\nabla}{2c} \quad \cdots\cdots(2)$$

Using the formula (2), we can solve it as scattering problem of one electron : an electron of energy E + ω is de-excited into a hole state of energy E with emission of photon and then it is re-excited to the upper level of E + ω again by absorbing a photon. The band structures of initial and final states are calculated as multiple scattering of the electron in the solid. In this formalism, we calculate only initial and final states which are connected with photoelectron wavefields outside of the solid. Therefore, computational time is saved a lot by using this formalism.

Theory of positronium formation at surface can be formalized in the similar way. Ps formation probability of direct process (one-electron process) due to an interaction V is given by

$$I(\vec{P}_{\parallel}, E_{ps}) = \sum \frac{L}{P_z} |<P_s, P_{\parallel}|V|\vec{q}>|\vec{k}>|^2 \quad \cdots\cdots(3)$$

where E_k is energy of initial electron in the solid, and q and k is the momentum of positron and electron respectively. $\epsilon_{kq,p}$ is the energy defference between initial and final state of the system defined by

$$\epsilon_{kq,p} = E_{ps}(\vec{p}) - E_k - E_q \quad \cdots\cdots(4)$$

Using the definition of Green's function,

$$|j,E><\rho(E)<j,E| = \frac{i}{2\pi}\sum_s \frac{|s,E_s><E_s,s|}{E-E_s+i\delta} - \frac{|s,E_s><s,E_s|}{E-E_s-i\delta}$$

$$= -\frac{1}{\pi} Im G_1^+(E) \quad \cdots\cdots(5)$$

we obtain the following formula :

$$I(\vec{P}_{\parallel}, E_{ps}) = -\frac{1}{\pi} Im \iiiint d\vec{r}_+ d\vec{r}_- d\vec{r}_+' d\vec{r}_-' \psi_{ps}(\vec{r}_+, \vec{r}_-) V(\vec{r}_+, \vec{r}_-) \varphi_q(\vec{r}_+)$$

$$\times G(\vec{r}_-, \vec{r}_-, E) \varphi_q^*(\vec{r}_+', \vec{r}_-') V(\vec{r}_+', \vec{r}_-') \psi_{ps}(\vec{r}_+', \vec{r}_-') \quad \cdots\cdots(6)$$

where ψ_{Ps} and φ_q is wave function of emitted Ps and incident positron. We can easily notice an analogy between eq. (6) and eq. (2). Thus, we can use most of all photoelectron calculation technique for the present Ps formation calculation. Positron corresponds to photoelectron and Ps corresponds to photon field.

Positron wavefield is calculated by LEED program with positron miffn-tin potential. According to the formalism of ref. 11, LEPD state inside the barrier is as follows ;

$$\phi_q(r) = L^{-1}\sum_g A_{jg}^+ exp[i\vec{K}_g^+ \cdot (\vec{r} - \vec{c}_j)] + A_{jg}^- exp[i\vec{K}_g^- \cdot (\vec{r} - \vec{c}_j)], \quad \cdots\cdots(7)$$

$$\vec{K}_g^{\pm}=(\vec{q}_{11}+\vec{g}, \pm\sqrt{2E_q-|\vec{q}_{11}+\vec{g}|^2}) \quad \cdots\cdots(8)$$

and outside the barrier,

$$\phi_q(\vec{r})=L^{-1}\sum_{\vec{g}} A_{1g}^{+} exp[i\vec{q}_g^{+}\cdot(\vec{r}-\vec{c}_1)]+A_{1g}^{-} exp[i\vec{q}_g^{-}\cdot(\vec{r}-\vec{c}_1)], \quad \cdots\cdots(9)$$

$$\vec{q}_g^{\pm}=(\vec{q}_{11}+\vec{g}, \pm q_z), \quad \cdots\cdots(10)$$

where the sum is done for receprocal lattice vectors on the surface as LEED calculation. The coefficients, $A^{\pm}_{jg}$ is calculated by LEPD calculation, where the suffix j means jth atomic layer from the top of the surface. Inside of the muffin-tin, the above plane wave should be re-written in the spherical wave expansion.

Positronium wavefield is presented below ;

$$\psi_{Ps}(\vec{r}_+, \vec{r}_-)=\frac{1}{L}\frac{1}{\sqrt{\pi a_0^3}} exp(-a_0|\vec{r}_+-\vec{r}_-|)$$

$$\times\left\{exp[i\vec{P}^{+}\cdot\frac{\vec{r}_++\vec{r}_-}{2}]+R\, exp[i\vec{P}^{-}\cdot\frac{\vec{r}_++\vec{r}_-}{2}]\right\} \quad \cdots\cdots(11)$$

where a_0 is the Bohr radius. Because of charge neutrality, we ignore multiple scattering effects due to muffin-tin for Ps. It would be true for metal, at least. However, definitely for ionic crystal, we should use a certain muffin-tin potential for Ps. Derivation of the muffin-tin potential of Ps itself would be an important and interesting topics, though we do not touch it here.

According to ref. 11, we assume that potential for Ps has the following from :

$$V_{Ps}(z)=\begin{cases} 0+i0 & (z<0) \\ -\phi_{Ps}+iV_i & (z>0) \end{cases} \quad \cdots\cdots(12)$$

where $z=\frac{1}{2}(z_+ + z_-)$ and $z=0$ is the position of the barrier. The solid is in $+z$ direction, the same as LEED theory [18], Though investigation for spacial shape of the barrier potential should be done in future, we here the above step potential for simplicity.

The interaction of Ps formation, V is approximately a Coulomb interaction between the

positron and an electron as derived in ref. 10.

$$V(\vec{r}_+,\vec{r}_-)=\frac{1}{|\vec{r}_+-\vec{r}_-|}e^{-\mu|\vec{r}_+-\vec{r}_-|} \quad \cdots\cdots(13)$$

We should point out that for 'on-shell' case, Ps formation matrix element due to eq. (13) is exactly same as the other Ps formation matrix due to image potentials as we showed in ref. 10. For energetic positron, we should use dynamical screening potential as in ref. 19. However, here , we use just the ordinary screened Coulomb potential for simplicity. In actual computational calculation, we could switch to the dynamical screening potential, because wave numbers and energies for electron, positron and Ps are all fixed for each multiple scattering calcultaions.

Hereafter we separate eq. (6) into the following two parts, according to ref. 11.

$$I=-\frac{1}{\pi}Im\int d\vec{r}_- H(\vec{r}_-)L(\vec{r}_-) \quad \cdots\cdots(14)$$

$$H(\vec{r}_-)=\int d\vec{r}_+\psi_{Ps}(\vec{r}_+,\vec{r}_-)\varphi_q(\vec{r}_+) \quad \cdots\cdots(15)$$

$$L(\vec{r}_-)=\int d\vec{r}_+'\int d\vec{r}_-' G(\vec{r}_-,\vec{r}_-',E_k)\varphi_q^*(\vec{r}_+')V(\vec{r}_+,\vec{r}_-)\psi_{Ps}(\vec{r}_+',\vec{r}_-') \quad \cdots\cdots(16)$$

3. Wavefield H($\vec{r}$) in muffin-tin potential

First, we reexpand H ($\vec{r}_-$) into sherical waves. To calculate transmission and reflection coefficients for a layer, we take a beam incident on the j-th layer

$$W_{j\vec{g}}^+ exp[iK_{1\vec{g}}^+\cdot(\vec{r}-\vec{c}_j)] \quad \cdots\cdots(17)$$

choosing as origin the center of an atom in the j-th layer. This plane wave results in spherical waves which in the absence of a potential would have the form near the atom at the origin,

$$\sum_{\ell m}A_{\ell m}^{(0)}(j)j_\ell(k,r)Y_{\ell m}(\theta,\varphi) \quad \cdots\cdots(18)$$

where

$$A^{(0)}_{1\ell m}(j)=\sum_{\vec{g}} 4\pi i^{\ell}(-1)^{m} W^{+}_{j\vec{g}} Y_{\ell -m}(\vec{K}^{+}_{1\vec{g}}) \quad \cdots\cdots(19)$$

and our shorthand notation for the argument of Y_{l-m} denotes the angular direction of $\vec{K}^{+}_{lg}$. Thus, for incident wave of amplitude W_j from $-z$ direction and V_j from $+z$ direction, we obtain

$$A^{(0)}_{2\ell m}(j)=\sum_{\vec{g}} 4\pi i^{\ell}(-1)^{m}[W^{+}_{j\vec{g}} Y_{\ell -m}(\vec{K}^{+}_{2\vec{g}})+V_{j\vec{g}} Y_{\ell -m}(\vec{K}^{-}_{2\vec{g}})] \quad \cdots\cdots(20)$$

According to ref. 17, multiple scattering between atoms within the layer modifies $A^{(0)}$:

$$A_{2\ell m}(j)=\sum_{2\ell' m'} A^{(0)}_{2\ell' m'}(j)(1-X_2)^{-1}_{\ell' m',\ell m} \quad \cdots\cdots(21)$$

where X_2 is presented in ref. 18 for a more detailed description of this calcultion. In our program code, X is calculated by XMATKC which is based on the method of Kambe [20]. So we have the expansion we seek about the atom at the origin in the j-th layer

$$\langle \phi|G_2|\vec{r}\rangle=\frac{1}{L}\sum_{\ell m} A_{2\ell m}(j)\psi_{2\ell}(E_2,|\vec{r}-\vec{c}_j|)Y_{\ell m}(\vec{r}-\vec{c}_j)e^{i\sigma_{2\ell}} \quad \cdots\cdots(22)$$

where ψ_{21} is the solution of the Schrodinger equation that is regular at the origin

$$\psi_{2\ell}=\frac{1}{2}(\psi^{+}_{2\ell}+\psi^{-}_{2\ell}) \quad \cdots\cdots(23)$$

and outside the atom reduces to

$$\psi_{2\ell}(E_2,r)=\frac{1}{2}[e^{i\sigma_{2\ell}}h^{(1)}_{\ell}(K_2 r)+e^{-i\sigma_{2\ell}}h^{(2)}_{\ell}(K_2 r)] \quad \cdots\cdots(24)$$

In rer. 11, the wavefield H (r) of eq. (15) is separated into four terms ;

$$H(\vec{r})=H_{11}(\vec{r})+H_{12}(\vec{r})+H_{21}(\vec{r})+H_{22}(\vec{r}) \quad \cdots\cdots(25)$$

Among them, waves in solid are H_{12} and H_{22}.

$$H_{12}(\vec{r}_-)=\frac{L^{-2}}{\sqrt{\pi a_0^3}}\sum_g\sum_\sigma\sum_\alpha R_0 D_1^{\sigma\alpha} exp[i(\vec{q}_{11g}^{\sigma}-\vec{p}_{11})\cdot\vec{r}_-]\, exp[(-2b_g-iq_{gz}^{\sigma})z_-]$$

$$+\frac{L^{-2}}{\sqrt{\pi a_0}^3}\sum_g\sum_\sigma TD_2^{\sigma} exp[i(\vec{q}_{11g}^{\sigma}-\vec{p}_{11})\cdot\vec{r}_-]$$

$$\times\left\{exp[(-b_g-ip_t^*/2)z_-]-exp[(-2b_g-iq_{gz}^{\sigma})z_-]\right\} \quad \cdots\cdots(26)$$

$$D_1^{\sigma\sigma}=A_{1g}^{\sigma}\frac{2\pi e^{-iq_{\sigma_{11}}\cdot c_1}}{b_g+iq_{gz}^{\sigma}-iq_z^{\sigma}/2)}, \quad \cdots\cdots(27)$$

$$D_2^{\sigma}=A_{1g}^{\sigma}\frac{2\pi e^{-iq_{\sigma_{11}}\cdot c_1}}{b_g(b_g+iq_{gz}^{\sigma}-ip_t^*/2)}, \quad \cdots\cdots(28)$$

$$b_g=(a^2+|q_g-p/2|^2)^{1/2} \quad \cdots\cdots(29)$$

$$\vec{q}_{\vec{g}}^{\pm}=(\vec{q}_{11}+\vec{g},\pm q_z) \quad \cdots\cdots(30)$$

$$a=a_0+\mu,\ R_+=1,\ R_-=R \quad \cdots\cdots(31)$$

$$H_{22}(\vec{r}_-)=\frac{L^{-2}}{\sqrt{\pi a_0}^3}\sum_g\sum_\sigma TD_3^{\sigma\alpha} exp[i(\vec{K}_{11g}^{\sigma}-\vec{p}_{11})\cdot\vec{r}_-]\left\{exp[(iK_{gz}^{\sigma}-p_t^*)z_-]\right.$$

$$\left.-exp[(-b_g-ip_t^*/2)z_-]\right\}+\frac{L^{-2}}{\sqrt{\pi a_0}^3}\sum_g\sum_\sigma TD_4^{\sigma} exp[i(K_{11g}^{\sigma}-p_{11})\cdot r_-]$$

$$\times\left\{exp[(b_g-ip_t^*/2)z_-]\, exp[(ik_{gz}^{\sigma}-ip_t^*/2-b_g)c]\right.$$

$$\left.-exp[i(K_{gz}^{\sigma}-p_t^*)z_-]\right\}, \quad \cdots\cdots(32)$$

$$D_3=A_{2g}^{\sigma}\frac{2\pi exp(-i\vec{K}_g^{\sigma}\cdot\vec{c}_1)}{b_g(iK_{gz}^{\sigma}-ip_t^*/2+b_g)}, \quad \cdots\cdots(33)$$

$$D_4=A_{2g}^{\sigma}\frac{2\pi exp(-i\vec{K}_g^{\sigma}\cdot\vec{c}_1)}{-b_g(iK_{gz}^{\sigma}-ip_t^*/2-b_g)}, \quad \cdots\cdots(34)$$

$$\vec{K}_g^{\pm}=(\vec{q}_{11}+\vec{g},\pm\sqrt{2E_q-|\vec{q}_{11}+\vec{g}|^2}) \quad \cdots\cdots(35)$$

Thus, we obtain for each wave numbers,

$$H_{12}+H_{22}=\frac{L^{-2}}{\sqrt{\pi a_0^3}}\sum_q\sum_\sigma\Big(\sum_\sigma R_\sigma D^{\sigma\sigma}-TD_2^\sigma\Big)e^{i(\vec{q}_{11}^{\,\sigma}g-\vec{p}_{11}),\vec{r}_-}e^{(-2bg-iq_{gz}^\sigma)z_-}$$

$$+\sum_g\sum_\sigma T(D_2^\sigma-D_3^\sigma)e^{i(\vec{q}_{11}g-\vec{p}_{11}),\vec{r}_-}e^{(-bg-i\frac{pt_1^*}{2})z_-}$$

$$+\sum_g\sum_\sigma T(D_3^\sigma-D_4^\sigma)e^{i(\vec{q}_{11}g-\vec{p}_{11}),\vec{r}_-}e^{i(K_{gz}^\sigma-pt_1^*)z}$$

$$+\sum\sum TD_4^\sigma e^{(iK_{gz}^\sigma z-i\frac{pt_1^*}{2}-bg)c}e^{i(\vec{q}_{11}g-\vec{p}_{11}),\vec{r}_-}e^{(bg-i\frac{pt_1^*}{2})z_-} \quad \cdots\cdots(36)$$

For atomic layers below the second, we should consider only terms including A_2.

From the above, we obtain coefficients W and V of eq. 20 for each wave numbers and obain $<\phi\ |\ G_2\ |\ \vec{r}>$ in spherical expansion.

4. Wavefield $L(\vec{r})$ in muffin-tin potential

Inside the atom (muffin-tin sphere), electronic Green's fuction is given by

$$G_{ij}(E,r,r')=-ik_1\sum_{\ell m}\psi_{1\ell}(r_<)\psi_{1\ell}^{+*}(r_>)Y_{\ell m}(\vec{r}'-\vec{c}_j)Y_{\ell m}^*(\vec{r}-\vec{c}_j) \quad \cdots\cdots(37)$$

where $r_<$ and $r_>$ means the smaller and the greater of $|\vec{r}-\vec{c}_j|$ and $\vec{r}'-\vec{c}_j|$. As shown above, the wave corresponds to photoelectron in ARUPS theory is given by

$$\langle\phi|G_2|\vec{r}\rangle=\frac{1}{L}\sum_{\ell m}A_{2\ell m}(j)\psi_{2\ell}(E_2,|\vec{r}-\vec{c}_j|)Y_{\ell m}(\vec{r}-\vec{c}_j)e^{i\sigma_{2\ell}} \quad \cdots\cdots(38)$$

Thus, if we assume that $\vec{r}'-\vec{c}_j$ is outside of the muffin-tin, we obtain,

$$\langle\vec{r}'|G_{1j}^{(0)+}G_2|\vec{r}\rangle=-i\frac{K_1}{L}\sum_{\ell m}\psi_{1\ell}^+(|\vec{r}'-\vec{c}_j|)Y_{\ell m}(\vec{r}'-\vec{c}_j|)\int d\vec{r}\,\psi_{1\ell}(|\vec{r}-\vec{c}_j|)Y_{\ell m}^*(\vec{r}-\vec{c}_j)$$

$$\times\sum_{\ell'm'}A_{2\ell'm'}^*(j)\psi_{2\ell'}^*(E_2,|\vec{r}-\vec{c}_j|)Y_{\ell'm'}^*(\vec{r}-\vec{c}_j)e^{-i\sigma_{2\ell'}}$$

$$=-i\frac{K_1}{L}\sum_{\ell m}\psi_{1\ell}^+(|\vec{r}'-\vec{c}_j|)Y_{\ell m}(\vec{r}'-\vec{c}_j)\sum_{\ell'm'}A_{2\ell'm'}^*(j)e^{-i\sigma_{2\ell'}}$$

$$\times\int\psi_{1\ell}(|\vec{r}-\vec{c}_j|)\psi^*_{2\ell'}(E_2,|\vec{r}-\vec{c}_j|)\,r^2dr$$

$$\times\int Y^*_{\ell m}(\vec{r}-\vec{c}_j)\,Y^*_{\ell' m'}(\vec{r}-\vec{c}_j)d\Omega \qquad \cdots\cdots(39)$$

where we use

$$\int Y^*_{\ell m}(\vec{r}-\vec{c}_j)Y^*_{\ell' m'}(\vec{r}-\vec{c}_j)d\Omega=(-1)^{-m}\delta_{\ell\ell'}\delta_{m-m'} \qquad \cdots\cdots(40)$$

which is obtained from

$$\int_o^\pi\sin\theta d\theta\int_o^{2\pi}d\varphi Y_{\ell' m'}(\theta,\varphi)Y_{\ell-m}(\theta,\varphi)(-1)^{\mathrm{m}}=\delta_{\ell\ell'}\delta_{\mathrm{mm}'} \qquad \cdots\cdots(41)$$

Therefore, we obtain

$$\langle\vec{r}'|G_{1j}G_2|\phi\rangle=-i\frac{K_1}{L}\sum_{\ell m}(-1)^{-\mathrm{m}}\psi^+_{1\ell}(|\vec{r}'-\vec{c}_j|)Y_{\ell m}(\vec{r}'-\vec{c}_j)A^*_{2\ell-m}(j)e^{-i\sigma_{2\ell}}$$

$$\times\int\psi_{1\ell}(|\vec{r}-\vec{c}_j|)\psi^*_{2\ell}(E_2,|\vec{r}-\vec{c}_j|)r^2dr \qquad \cdots\cdots(42)$$

In contrast to photoemission theory, we use only terms of $1=1'$, $\mathrm{m}=-\mathrm{m}'$.

Since the wavefield H is defined by eq. (15), it would be a problem to define ψ_{21}. From eq. (36), we should use four plane waves outside of the muffin-tin. Thus, we should solve Schrodinger equation for each waves to obtain $\psi_{21}{}^{\mathrm{a}}$, $\psi_{21}{}^{\mathrm{b}}$, $\psi_{21}{}^{\mathrm{c}}$, $\psi_{21}{}^{\mathrm{d}}$ by using electronic muffin-tin potential. We should get a sum for these four in eq. (42),

$$\langle\vec{r}'|G_{1j}G_2|\phi\rangle=-i\frac{K_1}{L}\sum_{\ell m}(-1)^{-\mathrm{m}}\psi^+_{1\ell}(|\vec{r}'-\vec{c}_j|)Y_{\ell m}(\vec{r}'-\vec{c}_j)A^*_{2\ell-\mathrm{m}}(j)e^{-i\sigma_{2\ell}}$$

$$\times\sum_{\alpha=a,b,c,d}\int\psi_{1\ell}(|\vec{r}-\vec{c}_j|)\psi^{\alpha*}_{2\ell}(E_2,|\vec{r}-\vec{c}_j|)\,r^2dr \qquad \cdots\cdots(43)$$

The obtained formula can be rewritten to the following form :

$$\langle\vec{r}'|G_{1j}G_2|\phi\rangle=\frac{1}{L}\sum_{\ell m}B^{(0)}_{1\ell m}(j)\psi^+_{1\ell}(|\vec{r}'-\vec{c}_j|)Y_{\ell m}(\vec{r}'-\vec{c}_j)e^{-i\sigma_{1\ell}} \qquad \cdots\cdots(44)$$

$$B^{(0)}_{1\ell m}(j)=-ik_1\sum_{\ell m}(-1)^m A^*_{2\ell-m}(j)e^{-i\sigma_{2\ell}}\int\psi_{1\ell}(|\vec{r}-\vec{c}_j|)\sum_{\alpha}\psi^{\alpha}_{2\ell}(E_2,\vec{r}-\vec{c}_j)r^2dr \quad \cdots\cdots(45)$$

which is just same as formula (96) in ref. 17. Since it is emitted wave from each atom, we should calculate multiple scattering of the wave $B_{1\ell m}{}^{(0)}$ within the layer.

It is easy to find amplitudes emitted by atoms other than the one at the origin : an atom removed a distance $\vec{R}$ from $\vec{c}_j$ emits wave-amplitudes

$$B^{(0)}_{1\ell m}\, exp[i(\vec{k}_{11}\cdot\vec{R}_{11})] \quad \cdots\cdots(46)$$

where

$$\vec{k}_{11}=\vec{q}_{11\vec{g}}-\vec{p}_{11} \quad \cdots\cdots(47)$$

Multiple scattering within the layer can be treated as same as ref. 17 and we obtain,

$$B_{1\ell m}=\sum_{\ell' m'}\frac{2B^{(0)}_{1\ell' m'}}{e^{2i\sigma_{1\ell'}}-1}[1-X(E,\vec{K}_{1_{11}})]^{-1}_{\ell' m',\ell m} \quad \cdots\cdots(48)$$

$B_{1\ell m}$ can be obtained by using subroutine BGPM in NEWPOOL or SLON with $B_{1\ell m}{}^{(0)}$ as input data.

The waves in eq. (48) are outgoing. However, according to ref. 17, we can easily use the same formalism we used above if we suppose that the outgoing waves were caused by incident waves,

$$\frac{1}{L}\sum_{\ell m}A^{(0)}_{1\ell m}j_\ell(K_1|\vec{r}'-\vec{c}_j|)Y_{\ell m}(\vec{r}'-\vec{c}_j) \quad \cdots\cdots(49)$$

where

$$A^{(0)}_{1\ell m}=\frac{2}{e^{2i\sigma_{1\ell}}-1}B^{(0)}_{1\ell m} \quad \cdots\cdots(50)$$

Substituting the above waves from the total wavefield, we obtain,

$$\frac{1}{L}\sum_{\ell m}[B_{1\ell m}-\frac{2B^{(0)}_{1\ell m}}{e^{2i\sigma_{1\ell}}-1}]\psi_{1\ell}Y_{\ell m}e^{i\sigma_{1\ell}}$$

$$+\frac{B^{(0)}_{1\ell m}}{e^{2i\sigma_{1\ell}}-1}[e^{i\sigma_{1\ell}}-e^{-i\sigma_{1\ell}}]\psi^{+}_{1\ell}Y_{\ell m} \quad \cdots\cdots(51)$$

where we use

$$2ij_\ell = h^{(0)}_\ell + h^{(2)}_\ell$$

$$= \psi^{+}_{1\ell}e^{-i\sigma_{1\ell}} + \psi^{-}_{1\ell}e^{i\sigma_{1\ell}}$$

$$= 2\psi_{1\ell}e^{i\sigma_{1\ell}} + \psi^{+}_{1\ell}(e^{-i\sigma_{1\ell}} - e^{i\sigma_{1\ell}}) \quad \cdots\cdots(52)$$

$$\psi = \frac{1}{2}(\psi^{+}+\psi^{-}) \quad \cdots\cdots(53)$$

Plane waves emitted from the layer are

$$\langle \vec{r}'|G^{(0)}_1 G_2|\phi\rangle = \sum_{\vec{g}} a^{+}_{j\vec{g}}\, e^{i\vec{k}^{+}_{1\vec{g}}\cdot(\vec{r}-\vec{c}_j)} \quad \cdots\cdots(Z>C_{jz})$$

$$= \sum_{\vec{g}} a^{-}_{j\vec{g}}\, e^{i\vec{k}^{-}_{1\vec{g}}\cdot(\vec{r}-\vec{c}_j)} \quad \cdots\cdots(Z<C_{jz}) \quad (54)$$

$$a^{\pm}_{j\vec{g}} = \sum_{\ell m}\sum_{\ell' m'} \frac{2\pi i}{K_1 \Omega k^{+}_{1gz}} \frac{2B^{(0)}_{1\ell' m'}(j)}{e^{2i\sigma_{1\ell'}}-1}(1-X_1)^{-1}_{\ell' m',\ell m} e^{i\sigma_{1\ell}}\sin(\delta_{1\ell}) i^{-\ell} Y_{\ell m}(\vec{K}^{\pm}_{1\vec{g}}) \quad \cdots\cdots(55)$$

Multiple scattering between layers is calculated as same as ref. 17. Namely, we define amplitudes of waves on the $-$z side of the j-th layer, travelling in the $+$z direction,

$$\frac{1}{L}\sum_{\vec{g}} d^{+}_{j\vec{g}}\, e^{i\vec{k}_{1\vec{g}}\cdot(\vec{r}-\vec{c}_j)} \quad \cdots\cdots(56)$$

and on the $+$z side travelling in the $-$z direction,

$$\frac{1}{L}\sum_{\vec{g}} d^{-}_{j\vec{g}}\, e^{i\vec{k}_{1\vec{g}}\cdot(\vec{r}-\vec{c}_j)} \quad \cdots\cdots(57)$$

we can calculate d_j to N-th layer by using the method presented in eq. (112) to eq. (118) of ref. 17. Computational calculation of this part is done in the subroutine PELOW of SLON.

The wave calculated above incident from both sides gives the incoming spherical waves about atoms in the layer. For the atoms at the origin of the j-te layer, corrected for multiple scattering in the usual way, the amplitudes are

$$F_{\ell'm'}=\sum_{\vec{g}}\sum_{\ell m}4\pi i^{\ell}(-1)^{m}[d_j^{+}Y_{\ell-m}(\vec{K}_{1\vec{g}}^{+})+d_{j\vec{g}}^{-}Y_{\ell-m}(\vec{K}_{1\vec{g}}^{-})](1-X_1)^{-1}_{\ell m,\ell'm'} \quad \cdots\cdots (58)$$

Adding the amplitudes originating within the j-th layer, the total wavefield outside the atom at the origin in the j-th layer is

$$\begin{aligned} L(\vec{r})&=\langle\vec{r}|G_1^{+}G_2|\phi\rangle \\ &=\frac{1}{L}\sum_{\ell m}[F_{\ell m}+B_{1\ell m}-\frac{2B_{1\ell m}^{(0)}}{e^{2i\sigma_1\ell}-1}]e^{i\sigma_{1\ell}}\psi_{1\ell}Y_{\ell m} \\ &\quad+\frac{B_{1\ell m}^{(0)}}{e^{2i\sigma_1\ell}-1}[e^{2i\sigma_{1\ell}}-e^{-i\sigma_{1\ell}}]\psi_{1\ell}^{+}Y_{\ell m} \quad \cdots\cdots (59)\end{aligned}$$

which is just same as eq. (120) of ref. 17 exept each coefficients, $F_{\ell m}$ and $B_{1\ell m}$.

5. Final coupling

For j-th layer, we define the H ($\vec{r}$) wave as follows,

$$\begin{aligned} H(\vec{r})&=H_{12}(\vec{r})+H_{22}(\vec{r}) \\ &=\frac{1}{L}\sum_{\substack{\alpha=a,b,\\ c,d}}\sum_{\ell'm'}A_{2\ell'm'}^{\alpha}(j)\psi_{2\ell'}^{\alpha}(E_2,|\vec{r}-\vec{c}_j|)Y_{\ell'm'}(\vec{r}-\vec{c}_j)e^{i\sigma_{2\ell'}} \quad \cdots\cdots (60)\end{aligned}$$

Substituting G_1^{+} has the effect of replacing ψ_1^{+} by $-\psi_1^{-}$, we obtain,

$$\begin{aligned} &-\frac{1}{\pi}Im\langle\phi|\psi_{ps}^{*}V\varphi_{+}G_1\varphi_{+}^{*}V\psi_{ps}|\phi\rangle \\ &=-\frac{1}{2\pi}Im\langle\phi|\psi_{ps}^{*}V\varphi_{+}(G_1^{+}-G_1^{-})\varphi_{+}^{*}V\psi_{ps}|\phi\rangle \quad \cdots\cdots (61)\end{aligned}$$

so that

$$\langle\vec{r}|\frac{1}{2}(G_1^+-G_1^-)V\varphi_+^*\psi_{ps}|\phi\rangle=\frac{1}{L}\sum_\alpha\sum_{\ell m}Z_{\ell m}^\alpha e^{i\sigma_{1\ell}}\psi_{1\ell}(|\vec{r}-\vec{c}_j|)Y_{\ell m}(\vec{r}-\vec{c}_j) \quad \cdots\cdots(62)$$

$$Z_{\ell m}^\alpha=F_{\ell m}^\alpha+B_{1\ell m}^\alpha+i\cot(\delta_{1\ell})e^{-2i\sigma_{1\ell}}B_{1\ell m}^{(0)\alpha} \quad \cdots\cdots(63)$$

Using eqs. (60) to (63) we find that

$$\begin{aligned}
I_j^\alpha&=-\frac{1}{\pi}Im\int H(\vec{r})L(\vec{r})d\vec{r}\\
&=-\frac{1}{\pi L^2}Im\sum_{\ell'm'}\sum_{\ell m}A_{2\ell'm'}^\alpha(j)e^{i\sigma_{2\ell'}}Z_{\ell m}^\alpha e^{i\sigma_{1\ell}}\\
&\quad\times\int d\vec{r}\,\psi_{2\ell'}^\alpha(E_2,|\vec{r}-\vec{c}_j|)\psi_{1\ell}(|\vec{r}-\vec{c}_j|)Y_{\ell'm'}(\vec{r}-\vec{c}_j)Y_{\ell m}(\vec{r}-\vec{c}_j)\\
&=-\frac{1}{\pi\Omega}Im\sum_{\ell'm'}\sum_{\ell m}A_{2\ell'm'}^\alpha(j)e^{i\sigma_{2\ell'}}Z_{\ell m}^\alpha e^{i\sigma_{1\ell}}\\
&\quad\times\int\psi_{2\ell'}^\alpha(|\vec{r}-\vec{c}_j|)\psi_{1\ell}(|\vec{r}-\vec{c}_j|)r^2dr\\
&\quad\times\int Y_{\ell'm'}(\vec{r}-\vec{c}_j)Y_{\ell m}(\vec{r}-\vec{c}_j)d\Omega\\
&=-\frac{1}{\pi\Omega}Im\sum_{\ell m}(-1)^{-m}A_{2\ell-m}^\alpha(j)e^{i\sigma_{2\ell}}Z_{\ell m}^\alpha e^{i\sigma_{1\ell}}\\
&\quad\times\int\psi_{2\ell}^\alpha(|\vec{r}-\vec{c}_j|)\psi_{1\ell}(|\vec{r}-\vec{c}_j|)r^2dr \qquad \cdots\cdots(64)
\end{aligned}$$

where we have summed contributions from each atom in the layer, and we have used

$$\psi_{1\ell}^*=\psi_{1\ell}$$

$$\delta_{1\ell}^*=\delta_{1\ell} \qquad \cdots\cdots(65)$$

The final expression for the current emitted into a beam with a parallel momentum p is

$$I_{MT}(\vec{P}_{11}, E_{PS})=\sum_{\alpha}\sum_{j} I_j^{(\alpha)} \qquad \cdots\cdots(66)$$

which complete our derivation. In the actual calculation, we should add the contribution of electron around a surface barrier, which has been presented by Ishii-Pendry [11],

$$I(\vec{P}_{11}, E_{PS})=I_{MT}(\vec{P}_{11}, E_{PS})+I_{IP}(\vec{P}_{11}, E_{PS}) \qquad \cdots\cdots(67)$$

6. Conclusion

We present formula of angle-resolved Ps formation spectrum for matirx element inside of muffin-tin potential. We can calculate every terms in the Ps formation theory by using the present result with the former result of Ishii and Pendry. To construct computational program code is easy if we use SLON as a base program cade.

Acknowledgement

The author is grateful for the Computer Center of the Institute for Molecular Science for the use of the HITAC M-680H and S-820/80 computer. The author also thank to Dr. S. Shindo of Tokyo-Gakugei University and Dr. Kawamura of Yamanashi University for valuable theoretical discussions.

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