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Relativistic Dynamical Theory of Electron Diffraction*

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The dynamical theory of electron diffraction is reformulated by solving Dirac's wave equation with a perturbation. The new scattering formula has the same form as Cowley and Moodie's (Acta Cryst. **10** 609 (1957)) and the present author's (J. Phys. Soc. Japan **14** (1959) 1513) at the nonrelativistic limit $c\to\infty$. The relativistic scattering formula, however, contains a correction factor due to the mass correction of electron, which has not so far been taken into account in the dynamical theory. This correction factor brings about modifications to some of the conclusions of the current theories. For instance, it is shown that the dynamical effect on the scattering does not vanish at the limit $\lambda\to 0$. Magnetic scattering of electrons by antiferromagnetic crystals is also discussed.

1. Introduction

In some recent experiments of electron diffraction, the electron beams having velocities comparable with that of light have been utilized. Although for such electrons the relativistic effect is expected to be far from negligible, the dynamical theory of electron diffraction based on nonrelativistic Schrödinger wave equation

$$\nabla^2\psi+K^2\psi+\frac{2m_0e}{\hbar^2}V\psi=0\,, \qquad (1)$$

has so far been applied only by taking account of the relativistic relation between wave-number K and accelerating voltage of electron. This theoretical procedure, however, cannot be correct, because it has been pointed out by Laue[1] that the *kinematical* scattering formula derived from Dirac's relativistic equation using Born approximation contains well-known Mott's correction factor[2]

$$1+\frac{\hbar^2K^2}{m_0^2c^2}\left(\frac{1+\cos\theta}{2}\right) \qquad (2)$$

$$(\theta:\ \textit{angle of scattering}),$$

corresponding to the increase of electron mass with the electron energy by the relativistic effect. We can expect naturally that the similar factors should be taken into account in formulae of the higher order approximations.

In the present work, the *dynamical* theory is reformulated by solving Dirac's wave equation with a perturbation method in order to study the relativistic form of the scattering formula and, besides, to discuss the scattering of electrons by periodic magnetic field.

2. The derivation of the scattering matrix

As is well known, Dirac's equation for an electron in an electromagnetic field $(\boldsymbol{A}, V)$ is given by

$$[\boldsymbol{\alpha}\cdot(c\boldsymbol{p}+e\boldsymbol{A})+\beta m_0c^2-eV]\psi=i\hbar\frac{\partial\psi}{\partial t}\,. \qquad (3)$$

We shall take

$$H' \equiv e\boldsymbol{\alpha}\boldsymbol{A}-eV$$

as a perturbation on the Hamiltonian for a free electron, and calculate the elements of the scattering matrix $\langle \boldsymbol{K}_h s|R|\boldsymbol{K}_0 s_0\rangle$, where $\boldsymbol{K}_0$ and $\boldsymbol{K}_h$ are the wave vectors of the incident and a scattered wave, respectively, and s_0 and s are the parameters distinguishing the spin states (and also the signs of energy) of electron.

We first consider the scattering of electrons by a periodic electrostatic field

* As a full report of this work appeared in the Journal of Phys. Soc. of Japan **16** (1961) 2226, the details of calculation should be referred to it.

$$V(r)=\begin{cases}\sum_{h} v_h \exp(2\pi i h r) & \text{for } 0\leq z\leq D\,,\\ 0 & \text{for } z<0,\ z>D\,.\end{cases}$$

In this case, the n-th order term of the scattering matrix can be written as

$$\langle \boldsymbol{K}_h s|R^{(n)}|\boldsymbol{K}_0 s_0\rangle=\left(\frac{e}{c^2\hbar^2}\right)^n\cdot E_{K_0}\sum_{g_{n-1}}\cdots\sum_{g_1}\frac{v_{h-(g_1+\cdots+g_{n-1})}\cdot v_{g_{n-1}}\cdots v_{g_2}\cdot v_{g_1}}{K_{h,z}\cdot K_{g_1+\cdots+g_{n-1},z}\cdots K_{g_1+g_2,z}\cdot K_{g_1,z}}$$

$$\times Z_n(\zeta_1,\zeta_2,\cdots,\zeta_{n-1},\zeta_h\,;D)\frac{1}{2^{n-1}}(u^*_{K_h}s\{E_{K_0}+H(\boldsymbol{K}_{g_1+\cdots+g_{n-1}})\}\cdots$$

$$\times\{E_{K_0}+H(\boldsymbol{K}_{g_1+g_2})\}\cdot\{E_{K_0}+H(\boldsymbol{K}_{g_1})\}u_{K_0}s_0)\,, \tag{4}$$

where

$$Z_n(\zeta_1,\zeta_2,\cdots,\zeta_{n-1},\zeta_h\,;D)$$

$$=\frac{1}{(2\pi)^n}\left[\sum_{m=1}^{n-1}\frac{\exp\{2\pi i(\zeta_m-\zeta_h)D\}-1}{\zeta_m(\zeta_m-\zeta_h)\prod\limits_{\substack{k=1\\k\neq m}}^{n-1}(\zeta_m-\zeta_k)}+(-1)^n\frac{\exp(-2\pi i\zeta_h D)-1}{\zeta_h\prod\limits_{k=1}^{n-1}\zeta_k}\right], \tag{5}$$

$$\left.\begin{aligned}&H(\boldsymbol{K}_{g_1})=c\hbar\boldsymbol{\alpha}\cdot\boldsymbol{K}_{g_1}+\beta m_0c^2\,,\\&\cdots\cdots\cdots\cdots,\\&\cdots\cdots\cdots\cdots,\\&H(\boldsymbol{K}_{g_1+\cdots+g_{n-1}})=c\hbar\boldsymbol{\alpha}\cdot\boldsymbol{K}_{g_1+\cdots+g_{n-1}}+\beta m_0c^2\,,\end{aligned}\right\} \tag{6}$$

$$E_{K_0}=+\sqrt{c^2\hbar^2K_0^2+m_0^2c^4}\,, \tag{7}$$

and $u_{K_0}s_0$ and $u_{K_h}s$ are Dirac's four component quantities (i.e. *spinors*). Concerning the details of the calculation and the definitions of the other quantities, reference may be made to the full report[3].

The formula (4) differs from the corresponding nonrelativistic formulae of Cowley and Moodie[4] and of the present author[5] by a factor

$$\left(\frac{1}{m_0c^2}\right)^n\cdot\frac{E_{K_0}}{2^{n-1}}\cdot(u^*_{K_h}s\{E_{K_0}+H(\boldsymbol{K}_{g_1+\cdots+g_{n-1}})\}\cdots\{E_{K_0}+H(\boldsymbol{K}_{g_1})\}u_{K_0}s_0)\,. \tag{8}$$

It can be easily shown that this factor approaches unity as $c\to\infty$, if $s=s_0$.

3. The discussion on the relativistic effect

Using Eq. (4), the transition probability from the incident wave $\boldsymbol{K}_0$ to a scattered wave $\boldsymbol{K}_h$ is given in the first approximation as

$$I^{(2)}_{h0}=\left(\frac{m_0e}{\hbar^2}\right)^2\cdot\frac{|v_h|^2}{K_{hz}^2}\cdot\frac{\sin^2(\pi\zeta_hD)}{(\pi\zeta_h)^2}\left[1+\frac{\hbar^2K_0^2}{m_0^2c^2}\left(\frac{1+\cos\theta_{0h}}{2}\right)\right], \tag{9}$$

where θ_{0h} is the angle between $\boldsymbol{K}_0$ and $\boldsymbol{K}_h$. As was already pointed out by Laue[1], the formula (9) is different from the nonrelativistic kinematical formula by Mott's factor which may be reduced to $1+(\hbar K_0/m_0c)^2$ since $\theta_{0h}\ll1$ as is usually the case. Similarly, the corresponding correction factors which appear in the second and the third order terms of transition probability are found to be

$$\left(1+\frac{\hbar^2K_0^2}{m_0^2c^2}\right)^{1/2}\cdot\left[1+\left(\frac{\hbar^2K_0^2}{m_0^2c^2}\right)\left(\frac{1+\cos\theta_{0h}+\cos\theta_{hg}+\cos\theta_{0g}}{4}\right)\right]\simeq\left(1+\frac{\hbar^2K_0^2}{m_0^2c^2}\right)^{3/2} \tag{10}$$

and

$$1+\frac{\hbar^2K_0^2}{m_0^2c^2}\left[1+\frac{1}{4}(\cos\theta_{hg'}+\cos\theta_{gh}+\cos\theta_{g'0}+\cos\theta_{g0})\right]$$

$$+\frac{1}{8}\left(\frac{\hbar^2K_0^2}{m_0^2c^2}\right)^2\Big(1+\cos\theta_{h0}+\cos\theta_{hg'}+\cos\theta_{gh}+\cos\theta_{0g'}+\cos\theta_{0g}+\cos\theta_{g'g}$$

$$+\cos\theta_{hg'}\cos\theta_{g0}+K_0^{-4}\cdot[\boldsymbol{K}_{g'},\boldsymbol{K}_h]\cdot[\boldsymbol{K}_g,\boldsymbol{K}_0]\Big)\simeq\left(1+\frac{\hbar^2K_0^2}{m_0^2c^2}\right)^2, \tag{11}$$

respectively, where $\boldsymbol{K}_g$ and $\boldsymbol{K}_{g'}$ are wave vectors of intermediate states. Using (8) we may easily obtain the general form of these correction factor as $[1+(\hbar K_0/m_0c)^2]^{n/2}$.

Since the factor $[1+(\hbar K_0/m_0c)^2]^{1/2}$ appears in the scattering formula always in the form $m_0\cdot[1+(\hbar K_0/m_0c)^2]^{1/2}$, and since

$$\sqrt{1+\frac{\hbar^2K_0^{\,2}}{m_0^{\,2}c^2}}=1\Big/\sqrt{1-\left(\frac{v}{c}\right)^2}\quad (v\text{: velocity of electron}),$$

the correction factors explained above should be interpreted as those due to the relativistic correction to the electron mass. Thus, it has been proved clearly that the relativistic dynamical theory of electron diffraction may be obtained *with sufficient accuracy* by starting from an equation

$$\nabla^2\psi+K^2\psi+\frac{2m_0e}{\hbar^2}\left(1+\frac{\hbar^2K^2}{m_0^{\,2}c^2}\right)^{1/2}V\psi=0. \tag{12}$$

The various experimental aspects of the relativistic effect are reported in another paper by Miyake, Fujiwara and Suzuki[6] in this volume. As an important example, however, we shall discuss the dependence of the reflexion intensity on wavelength. Since, in the current theory, the scattering matrix may be expanded as a power series in $m_0\lambda$, it follows that the higher order terms (i.e. *dynamical terms*) vanish and only the first order term (i.e. *kinematical term*) remains at the limit $\lambda\to 0$. In the relativistic theory, however, the series in $m_0\lambda$ must be replaced by that in $m_0\lambda\cdot[1+(h/m_0c\lambda)^2]^{1/2}$. Since, as shown in Fig. 1, $\lambda\cdot[1+(h/m_0c\lambda)^2]^{1/2}$ approaches not zero but a finite value $h/m_0c=0.0242$Å (*Compton wavelength of electron*) as $\lambda\to 0$, the dynamical terms no longer vanish at this short-wavelength limit. In other words, *the kinematical condition cannot be realized by raising the energy of electron.* This conclusion may be well illustrated by pointing out, for example, that the gap between doubled dispersion surfaces in the conventional dynamical theory approaches a finite minimum value as the energy increases.

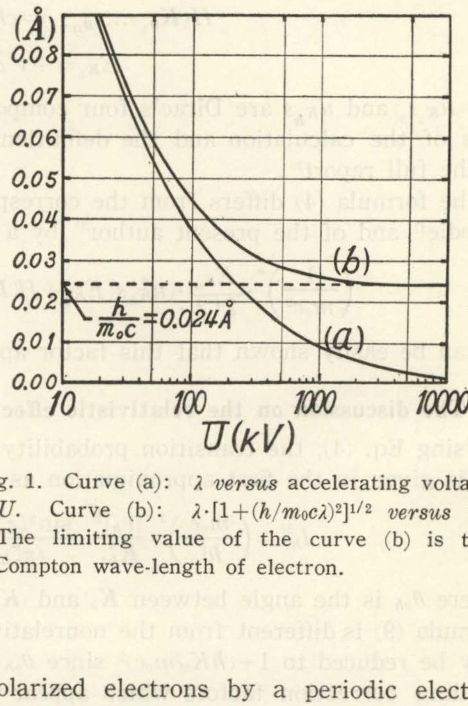

Fig. 1. Curve (a): λ *versus* accelerating voltage U. Curve (b): $\lambda\cdot[1+(h/m_0c\lambda)^2]^{1/2}$ *versus* U. The limiting value of the curve (b) is the Compton wave-length of electron.

4. The magnetic scattering of electrons

We shall consider the scattering of perfectly polarized electrons by a periodic electromagnetic field which is given by scalar and vector potentials,

$$V(r)=\begin{cases}\sum\limits_h v_h\exp(2\pi i h r) & \text{for } 0\le z\le D,\\ 0 & \text{for } z<0,\ z>D,\end{cases}$$

and

$$\boldsymbol{A}(r)=\begin{cases}\sum\limits_h \boldsymbol{A}_h\exp(2\pi i h r) & \text{for } 0\le z\le D,\\ 0 & \text{for } z<0,\ z>D.\end{cases}$$

The transition probability from $\boldsymbol{K}_0$ to $\boldsymbol{K}_h$ is written in the first approximation as

$$I_{h0}=\left(\frac{m_0 e}{\hbar^2}\right)^2\cdot\frac{1}{K_{h,z}^2}\cdot\frac{\sin^2(\pi\zeta_h D)}{(\pi\zeta_h)^2}\cdot\left[|v_h|^2\cdot\left(1+\frac{\hbar^2 K_0^2}{m_0^2c^2}\cdot\frac{1+\cos\theta_{0h}}{2}\right)\right.$$
$$-\frac{\hbar}{m_0^2c^3}\cdot E_{K_0}\cdot(\boldsymbol{K}_h+\boldsymbol{K}_0)\cdot\mathrm{Re}\,(v_h\boldsymbol{A}_h^*)$$
$$+\frac{\hbar^2}{2m_0^2c^2}\{(\boldsymbol{K}_0\cdot\boldsymbol{K}_0-\boldsymbol{K}_h)(\boldsymbol{A}_h^*\boldsymbol{A}_h)+2\mathrm{Re}\,((\boldsymbol{A}_h^*\boldsymbol{K}_h)(\boldsymbol{A}_h\boldsymbol{K}_0))\}$$
$$\mp\left\{\frac{\hbar^3K_{0z}}{m_0^2c(m_0c^2+E_{\ 0})}\cdot[\boldsymbol{K}_0,\boldsymbol{K}_h]\cdot\mathrm{Im}\,(v_h\boldsymbol{A}_h^*)+\frac{\hbar}{m_0c}\cdot[\boldsymbol{K}_h-\boldsymbol{K}_0,\mathrm{Im}\,(v_h\boldsymbol{A}_h^*)]_z\right.$$
$$\left.\left.+\frac{\hbar^2K_{0z}}{m_0^2c^2}\cdot(\boldsymbol{K}_h-\boldsymbol{K}_0)\cdot[\mathrm{Im}\,(\boldsymbol{A}_h),\mathrm{Re}\,(\boldsymbol{A}_h)]\right\}\right], \tag{13}$$

where minus or plus in the double sign should be taken according as the incident beam is polarized in the direction parallel or antiparallel to the z-axis, respectively. Therefore, when the incident beam is unpolarized, the fourth them in the square bracket vanishes by cancellation.

The above assumption that the vector potential $\boldsymbol{A}(\boldsymbol{r})$ in crystal has the periodicity of crystal lattice may be justified at least for antiferromagnetic substances, and, for such substances, the general expression of the Fourier component has been given by Miyake[7] as

$$\boldsymbol{A}_h=i2\mu_B\cdot\rho_h^m[\boldsymbol{h},\boldsymbol{M}]/|\boldsymbol{h}|^2\,, \tag{14}$$

where μ_B is Bohr magneton and $\rho_h^m\cdot\boldsymbol{M}$ is the Fourier component of the net density of the magnetic moments of electrons. Using Eqs. (13) and (14), we may easily show that the ratio of the magnetic scattering (i.e. the third term in the square bracket in (3)) to the electrostatic one (i.e. the first term) cannot exceed

$$|\boldsymbol{A}_h|^2/|v_h|^2\simeq10^{-4}\cdot|F_h^m/F_h|^2\,,$$

where F_h is the structure amplitude in the ordinary sense, and F_h^m is that for magnetic electrons only.

This paper was read at the Meeting of the Physical Society of Japan which was held in March 31st in 1961. In that talk, some comments of the experimental aspects of relativistic effect worked by Prof. Miyake and myself[6] were also quoted. A similar talk was given in more detail also in an informal meeting held in April at Nagoya University before Nagoya and Kyoto groups.

In conclusion the author wishes to express his sincere thanks to Prof. Shizuo Miyake for his kind guidance and encouragement throughout the present work.

References

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DISCUSSION

J. M. Cowley: Am I correct in supposing that, since the geometric term in the n-beam relativistic expression is not modified by relativistic effects, the Ewald sphere becomes planar as λ tends to zero and only the modified structure factor terms remain, giving a sort of phase-grating approximation?

K. Fujiwara: I think, it is probably so. But, we must notice that the maximum foil thickness, to which the phase-grating approximation is applicable, decreases because of the increase of dynamical interactions caused by the relativistic effect.

M. J. WHELAN : At Cambridge we have been interested for some time in the energy dependence of absorption process in metal foils, and since after I left England four months ago, Dr. Howie in our laboratory has recently formulated, independently of Dr. Fujiwara, the problem using Dirac's equation and has arrived at similar conclusions to those of Dr. Fujiwara. He has asked me to read the following coutribntion on his behalf.

We start from Dirac's equation

$$\{i\hbar c\boldsymbol{\alpha}\cdot\nabla-(W+V)\}\psi=\beta m_0c^2\psi\,, \tag{1}$$

where W is the total energy of the electron in the potential $V(r)$, i.e. $W=m_0c^2/\sqrt{1-v^2/c^2}$, m_0 is the rest mass of the electron, v is the electron velocity. ψ is the 4-component Dirac wave function, ψ_λ, $\lambda=1, 2, 3, 4$, corresponding to the two spin states of positive and negative total energy. $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ are the Dirac matrices. We operate on equation (1) from right with $\{i\hbar c\boldsymbol{\alpha}\nabla+(W+V)\}$ and use the relations $\boldsymbol{\alpha}_x\boldsymbol{\alpha}_y+\boldsymbol{\alpha}_y\boldsymbol{\alpha}_x=0$, $\boldsymbol{\alpha}_x^2=\boldsymbol{\alpha}_y^2=\boldsymbol{\alpha}_z^2=\boldsymbol{\beta}^2=1$, $\boldsymbol{\alpha}_x\boldsymbol{\beta}+\boldsymbol{\beta}\boldsymbol{\alpha}_x=0$ etc., and we obtain the equation

$$\nabla^2\psi_\lambda+\left[4\pi^2\chi^2-\frac{2WV}{\hbar^2c^2}+\frac{V^2}{\hbar^2c^2}-\frac{i}{\hbar c}\boldsymbol{\alpha}\cdot(\nabla V)\right]\psi_\lambda=0\,, \tag{2}$$

$\lambda=1, 2, 3, 4$. $4\pi^2\chi^2=(W^2-m_0^2c^2)/h^2c^2$, χ is the relativistic wave vector,

$$\chi=\frac{m_0v}{\sqrt{1-v^2/c^2}}\cdot\frac{1}{2\pi\hbar}\,.$$

We put $V=-\sum V_h\exp(2\pi i\boldsymbol{h}\cdot\boldsymbol{r})$, $\psi_\lambda=\sum_h\phi_h^{(\lambda)}\exp\{2\pi i(\boldsymbol{k}_0+\boldsymbol{h})\cdot\boldsymbol{r}\}$, and neglect the term in V^2 since $V\ll W$. In the two-beam approximation taking z normal to g (i.e. $g_z=0$, i.e. $\partial V/\partial z=0$) we obtain the fundamental equation of relativistic theory

$$\begin{bmatrix} (\Lambda^2-k_0^2) & U_{-g} & 0 & 0 & 0 & 0 & 0 & \eta^*_{-g} \\ U_g & (\Lambda^2-k_g^2) & 0 & 0 & 0 & 0 & \eta^*_g & 0 \\ 0 & 0 & (\Lambda^2-k_0^2) & U_{-g} & 0 & \eta_{-g} & 0 & 0 \\ 0 & 0 & U_g & (\Lambda^2-k_g^2) & \eta_g & 0 & 0 & 0 \\ 0 & 0 & 0 & \eta_{-g} & (\Lambda^2-k_0^2) & U_{-g} & 0 & 0 \\ 0 & 0 & \eta_g & 0 & U_g & (\Lambda^2-k_g^2) & 0 & 0 \\ 0 & \eta_{-g} & 0 & 0 & 0 & 0 & (\Lambda^2-k_0^2) & U_{-g} \\ \eta_g & 0 & 0 & 0 & 0 & 0 & U_g & (\Lambda^2-k_g^2) \end{bmatrix} \begin{bmatrix} \phi_0^{(1)} \\ \phi_g^{(1)} \\ \phi_0^{(2)} \\ \phi_g^{(2)} \\ \phi_0^{(3)} \\ \phi_g^{(3)} \\ \phi_0^{(4)} \\ \phi_g^{(4)} \end{bmatrix} = 0 \tag{3}$$

where

$$U_g=\frac{WV_g}{2\pi^2\hbar^2c^2}=\frac{m_0}{2\pi^2\hbar^2}\frac{V_g}{\sqrt{1-v^2/c^2}}\,, \tag{4}$$

$$\Lambda^2=\chi^2+\frac{WV_0}{2\pi^2\hbar^2c^2}\,, \tag{5}$$

$$\eta_g=\frac{-(g_x+ig_y)V_g}{2\pi\hbar c}=\frac{-(g_x+ig_y)U_g\pi\hbar c}{W}\,, \tag{6}$$

Λ is the mean wave vector after correction for the inner potential. Dr. Howie points out that the relativistic correction factor $\sqrt{1-v^2/c^2}$ to the U_g term enters equation (4) in agreement with Dr. Fujiwara's result.

Strictly to diagonalize the matrix of equation (3) we would have to take account of the cross diagonal terms η_g. Thus there will be a slightly different dispersion surface for each of 4 components of the wave function. The effect of the terms is to cause some uncertainty about spin for a wave diffracted through an angle ϑ. η_g is always associated with spin-flips and diffraction. However, in practice $|\eta_g/v_g|\sim1/50\cdot(\chi\pi\hbar c/W)$

$=1/100 \cdot v/c$, i.e. about 1%, and decreases with increasing energy. Since none of the η_g appears on the main diagonal, the correction to the eigenvalues (k_0, k_g) in equation (3) is second order in η_g, i.e. about 0.01%. Thus to a good approximation the 4 Dirac wave functions are described by the usual two-beam wave functions. Dr. Howie points out that the treatment can readily be extended to the multiple-beam case (the η_g terms appear in more places of course) provided (a) we use a relativistic value of wave vector χ, and (b) the U_g terms are corrected by the $\sqrt{1-v^2/c^2}$ factor. This is equivalent to correction of the mass in Schrödinger's non-relativistic equation, a result identical with that of Dr. Fujiwara.

We can apply these results to geometrical theories of the dynamical theory (see references cited in my paper in this Volume).

Dr. Howie also points out that extinction distance $\xi_g(=\chi/U_g)$ should vary as $hc/2V_g(v/c)$, and that in the Yoshioka's general theory of inelastic scattering we should expect to have to correct the H' terms in his theory by the factor $\sqrt{1-v^2/c^2}$. Since these terms appear as products in the expressions for C^i_{00} and C^i_{g0}, it follows that the absorption parameters ξ'_0 and ξ'_g (notation of Whelan's paper in this Volume) vary as $(v/c)^2$. I believe Dr. Hashimoto has some experimental results at different kilo-voltages.

K. FUJIWARA : Has Dr. Howie's work been already published?

M. J. WHELAN : No. His work was not done when I left England.

P. B. HIRSCH : I think it is noticiable that, as Dr. Howie has shown, the extinction distance is proportional to the velocity of the electrons, and that the (absorption coefficient)$^{-1}$ is proportional to the (velocity)^2.

It is interesting to observe that the extinction distance reaches a limit which has a value 1.8 times the extinction distance at 100 kV.

S. MIYAKE : On behalf of Dr. Fujiwara, I should like to add that his treatment of the relativistic dynamical theory is quite rigorous, and it should particularly be noticed the fact that the relativistic factors contain, as he has derived, terms relating to scattering angles, and he showed that they can be approximated to the form of the multiples of $(1+h^2/m_0{}^2c^2\lambda^2)^{1/2}$.

R. UYEDA : I'd like to just mention that the voltage-dependence of Yoshioka's terms including relativistic effect was already studied quantitatively by Prof. Miyake's laboratory, as will be reported in this Symposium by himself.