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On a mosaic graphite spectrometer without collimators¹

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An X-ray spectrometer was constructed for the detection of very weak lines having a large energy shift from the primary (Rayleigh) line. The spectrometer has no collimators and uses a graphite mosaic crystal as an analyser. The dependence of the resolution and intensity on one another is examined and both are calculated as a function of the scatterer-analyser distance. The spectrometer was tested in X-ray inelastic scattering experiments.

On a construit un spectromètre à rayons X pour la détection de raies très faibles séparées par un décalage d'énergie considérable de la raie primaire (Rayleigh). Ce spectromètre n'a pas de collimateurs et utilise comme analyseur un cristal de mosaïque de graphite. On examine la façon dont la résolution et l'intensité dépendent l'une de l'autre, et on calcule l'une et l'autre en fonction de la distance diffuseur-analyseur. On a fait l'essai de ce spectromètre dans des expériences de diffusion inélastique des rayons X.

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I. Introduction

In a spectrometry experiment, the two main factors which must be examined are the resolution and the intensity provided by the instrumentation. For lines close to a strong primary line (they result for example from Compton or plasmon scattering) the resolution is more important than the intensity. On the other hand, for weak excitations with a large energy shift from the primary line (e.g., Raman effect), high intensity is the important factor.

In a spectrometer using as an analyser a perfect crystal, all the rays from a point source after reflection are focused at the same point on a circumference containing the source and the rotation point of the crystal (focusing circle). The above focusing effect is not realized in a practical application because the analyser has a mosaic spread, the primary line has a natural width, and the dimensions of the source cannot be neglected.

In the present paper a spectrometer is described that uses a flat mosaic graphite crystal as an analyser and an extended flat scatterer as a secondary X-ray source. It is important to examine the resolution and the intensity that can be achieved with such a contrivance and the relationship of these properties to one another.

II. The resolution versus the scatterer-analyser distance

(i) *Form of a monochromatic line after reflection on the analyser due to its mosaic spread*

If the mosaic spread of the analyser causes a broad-

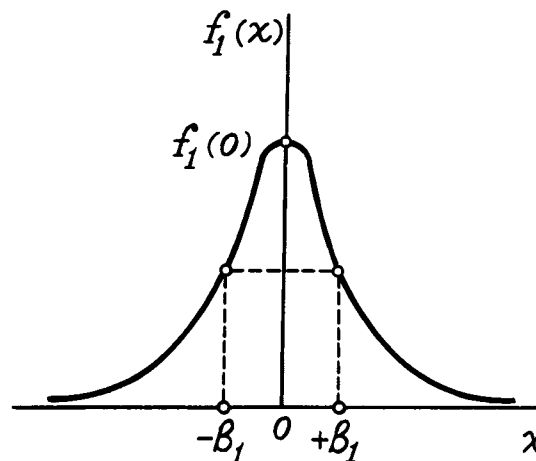


FIG. 1. Form of the line due to the mosaic spread of the analyser.

ening $2\beta_1$ (Fig. 1), the form of a strictly monochromatic line after reflection on the analyser can be expressed approximately by a Gaussian function (1):

$$f_1(x) = f_1(0)e^{-kx^2}$$

where $x = \vartheta - \vartheta_{\text{Bragg}}$. The conditional relation

$$f_1(\beta_1) = \frac{1}{2}f_1(0)$$

leads to

$$[1] f_1(x) = f_1(0)e^{-(\ln 2)\beta_1^{-2}x^2}$$

(ii) *Form of the line due to its natural width*

The natural width of a Gaussian primary line produces a broadening $2\beta_2$, and the function $f_2(x)$ of the line will be similar to [1]:

$$f_2(x) = f_2(0)e^{-(\ln 2)\beta_2^{-2}x^2}$$

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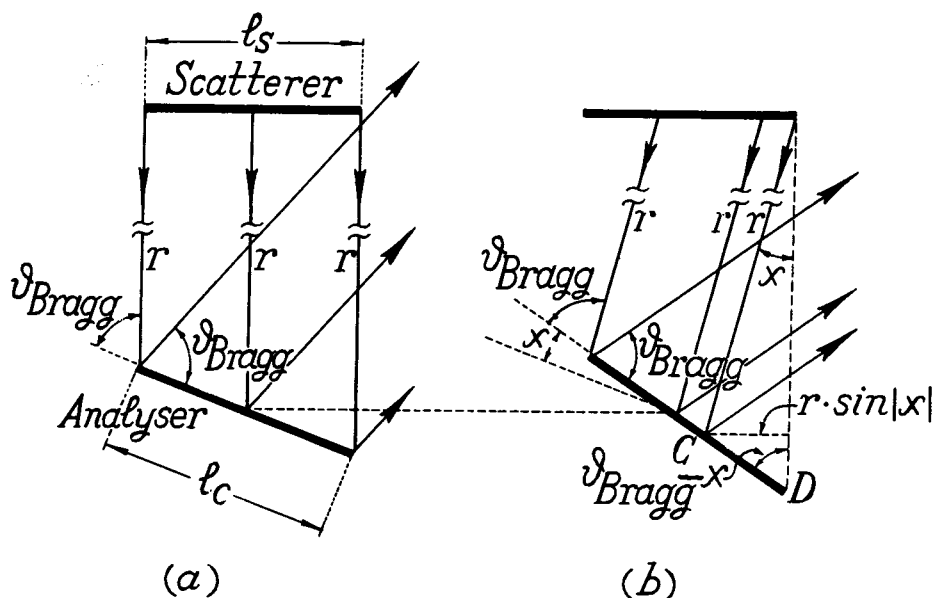


FIG. 2. Configuration of the scatterer and analyser for the determination of the line shape due to the scatterer dimensions.

(iii) *Combination of the mosaic spread and the natural width broadening*

The form of the line due to both broadenings will be (2):

$$f(x) = f_1(0)f_2(0)(\pi / \ln 2)^{1/2}(\beta_1^{-2} + \beta_2^{-2})^{1/2} \times e^{-(\ln 2)(\beta_1^2 + \beta_2^2)^{-1}x^2}$$

Introducing new variables

$$\beta^2 = \beta_1^2 + \beta_2^2$$

$$\frac{1}{2}z^2 = (\ln 2)\beta^{-2}x^2$$

and putting

$$\varphi(z) = \frac{1}{\sqrt{2\pi}} e^{-z^2/2}$$

we have

$$[2] f(z) = f(0)\varphi(z)$$

(iv) *Form of the line due to the dimensions of the scatterer*

We consider the case of a strictly monochromatic radiation and a perfect crystal as analyser. For a very long scatterer-analyser distance in comparison with their dimensions, we can assume that all the rays from the scatterer to the analyser are approximately parallel, of equal length r , and on the meridian of the apparatus. Figure 2a shows the analyser in the proper position to satisfy Bragg's law. In Fig. 2b the analyser has been rotated by an angle:

$$|x| = |\vartheta - \vartheta_{\text{Bragg}}|$$

In this case, a certain part,

$$(CD) \approx \frac{r \sin |x|}{\sin (\vartheta - |x|)} \approx \frac{r|x|}{\sin \vartheta_{\text{Bragg}}}$$

of the analyser cannot be irradiated under the Bragg angle and does not contribute to the intensity of the line.

Considering the intensity proportional to the effective length:

$$l_c - (CD) \approx l_c - \frac{r|x|}{\sin \vartheta_{\text{Bragg}}} = l_c \left[1 - \frac{|x|}{l_c/r \sin \vartheta_{\text{Bragg}}} \right]$$

we will have for the profile of the line the analytical expression:

$$g(x) = g(0) \left[1 - \frac{|x|}{l_c/r \sin \vartheta_{\text{Bragg}}} \right]$$

Introducing the parameter

$$\gamma = (2 \ln 2)^{1/2} \frac{1}{\beta} \frac{l_c}{r} \sin \vartheta_{\text{Bragg}}$$

the form of the line is expressed by

$$[3] g(z) = g(0) \left[1 - \frac{|z|}{\gamma} \right]$$

which is equivalent to the equations

$$g(z) = g(0) \left[1 + \frac{z}{\gamma} \right], \quad -\gamma \leq z \leq 0$$

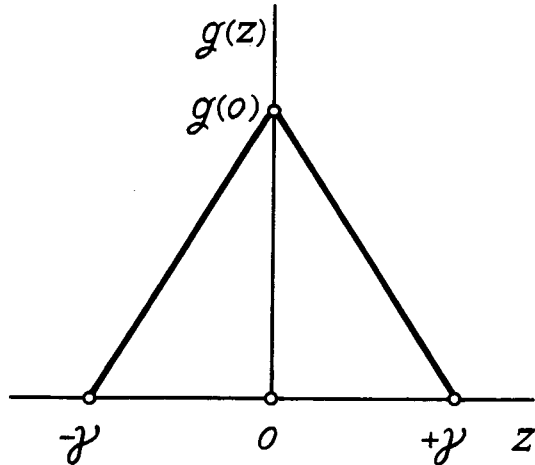


FIG. 3. Form of the line due to the scatterer dimensions.

$$g(z) = g(0) \left[1 - \frac{z}{\gamma} \right], \quad 0 \leq z \leq \gamma$$

The line form described by [3] is plotted in Fig. 3.

(v) *Form of the line due to the total broadening*

The final profile of the line is the convolution (2) of [2] and [3]:

$$I(z) = \int_u f(u)g(z-u) du$$

$$= \frac{1}{\sqrt{2\pi}} f(0)g(0) \int_u e^{-u^2/2} \left[1 - \frac{|z-u|}{\gamma} \right] du$$

The limits of the integral are determined from the conditions

$$1 - \frac{|z-u|}{\gamma} = \begin{cases} 1 & \text{for } z = u \\ 0 & \text{for } |z-u| \geq \gamma \end{cases}$$

which yield

$$I(z) = \frac{1}{\sqrt{2\pi}} f(0)g(0) \left[\int_{z-\gamma}^z e^{-u^2/2} \left[1 - \frac{z-u}{\gamma} \right] du + \int_z^{z+\gamma} e^{-u^2/2} \left[1 + \frac{z-u}{\gamma} \right] du \right]$$

Putting

$$\Phi(u) = \frac{1}{\sqrt{2\pi}} \int_u e^{-u^2/2} du$$

we conclude

$$[4] \quad I(z) = \frac{1}{\gamma} f(0)g(0) [(z+\gamma)\Phi(z+\gamma) + (z-\gamma)\Phi(z-\gamma) - 2z\Phi(z) + \varphi(z+\gamma) + \varphi(z-\gamma) - 2\varphi(z)]$$

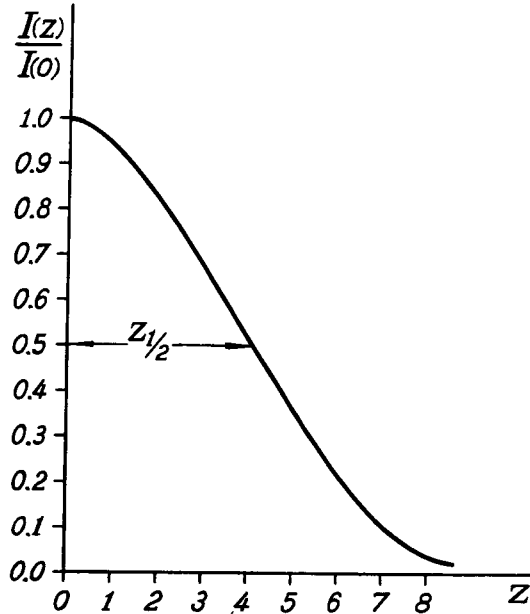


FIG. 4. The ratio $I(z)/I(0)$ versus z , for $r = 125$ cm.

and

$$I(0) = \frac{2}{\gamma} f(0)g(0) [\gamma\Phi(\gamma) + \varphi(\gamma) - \varphi(0)]$$

(vi) *The full width at the half maximum ($\Delta\vartheta_{FWHM}$)*

The analytical determination of $\Delta\vartheta_{FWHM}$ from [4] is not practical, so we determine it as follows.

We calculate the function $I(z)/I(0)$ versus r for the concrete case of a spectrometer with the following parameters. The analyser is a flat mosaic graphite crystal with $l_c = 3$ cm length and 2.5 cm height which has a mosaic spread (3):

$$2\beta_1 = 0.4^\circ = 6.98 \times 10^{-3} \text{ rad}$$

Radiation CrK_β is reflected on the (0006) planes of the analyser at an angle of $\vartheta_{\text{Bragg}} = 68.81^\circ$. The natural width of the CrK_β line at the half maximum is (4)

$$2\beta_2 = 4.14 \text{ eV}$$

The resolving power 4.33×10^{-4} rad/eV of the present spectrometer gives

$$2\beta_2 = 1.36 \times 10^{-3} \text{ rad}$$

Consequently

$$\beta = 3.56 \times 10^{-3} \text{ rad}$$

Figure 4 shows the ratio $I(z)/I(0)$ versus z for $r = 125$ cm. From similar curves for various r , we find $z_{1/2}$ and calculate the $\Delta\vartheta_{FWHM}$ and the resolution S from the relations

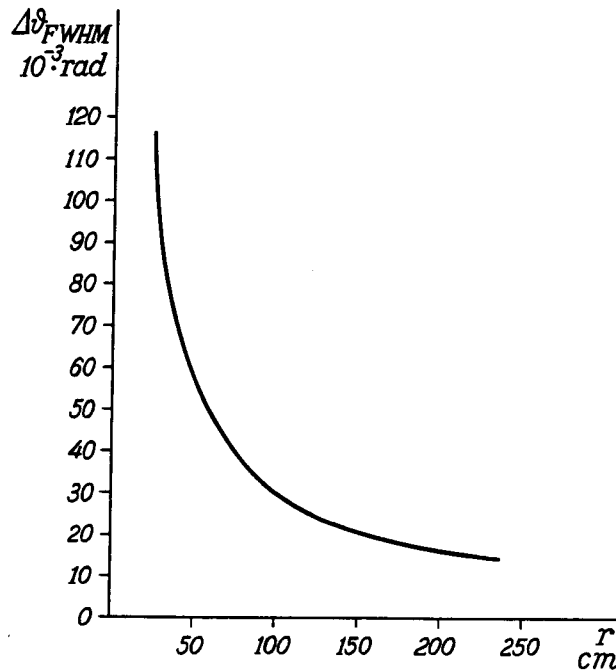


FIG. 5. Full width at half maximum versus r .

$$\Delta\vartheta_{FWHM} = 2x_{1/2} = \frac{2\beta}{\sqrt{2 \ln 2}} z_{1/2}$$

$$S = \frac{\tan \vartheta_{Bragg}}{\Delta\vartheta_{FWHM}}$$

Figures 5 and 6 show the $\Delta\vartheta_{FWHM}$ and S versus r .

The large values of $\Delta\vartheta_{FWHM}$ show that the spectrometer described is not suitable when discriminating lines are close to each other, such as the Compton and plasmon excitations in relation to the primary line.

III. The intensity versus scatterer-analyser distance

We consider the infinitesimal surfaces $dS_s = dh_s dl_s$ of the scatterer and $dS_c = h_c dl_c$ of the analysing crystal (Fig. 7). The solid angle $d\Omega$ of the rays from the center of dS_s to dS_c is

$$d\Omega = \frac{dS_c \sin \vartheta_{Bragg}}{r^2} = \frac{h_c dl_c \sin \vartheta_{Bragg}}{r^2}$$

The number of photons emitted within the solid angle $d\Omega$ from the surface dS_s will be

$$dA_s = M dS_s d\Omega = M dh_s dl_s \frac{h_c \sin \vartheta_{Bragg} dl_c}{r^2}$$

where M is a coefficient that depends on the scatterer. From Fig. 7 we have

$$\frac{dl_c}{\sin(d\vartheta)} = \frac{r}{\sin(180^\circ - \vartheta_{Bragg})}$$

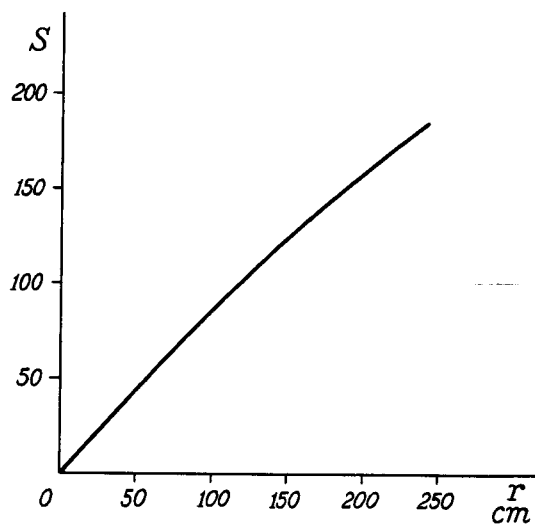


FIG. 6. The resolution S versus r .

or

$$dl_c = \frac{r}{\sin \vartheta_{Bragg}} d\vartheta$$

Therefore, the number of photons emitted from the whole surface of the scatterer towards the surface dS_c of the analyser will be

$$dA_s = \frac{M}{r} h_c h_s l_s d\vartheta$$

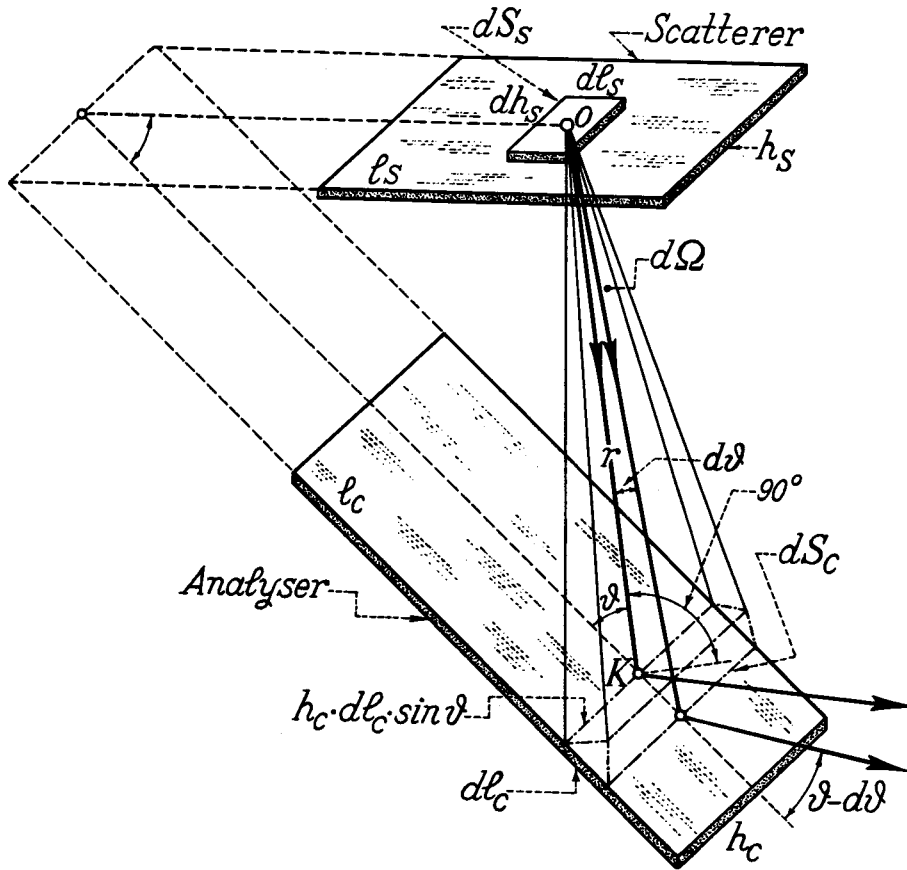


FIG. 7. Configuration of the scatterer and analyser for the calculation of the scattered intensity.

A part, dA_c , of these photons is reflected by the analyser and

$$dA_c = \rho(\vartheta) dA_s$$

where $\rho(\vartheta)$ is its reflectivity. For the analyser set at the Bragg position, the total number of photons/s reflected from the whole surface will be

$$[5] \quad A_c = \frac{M}{r} h_c h_s l_s \int_{\vartheta_{\text{Bragg}} - \epsilon}^{\vartheta_{\text{Bragg}} + \epsilon} \rho(\vartheta) d\vartheta$$

where $\vartheta_{\text{Bragg}} - \epsilon$ and $\vartheta_{\text{Bragg}} + \epsilon$ are the limiting angles beyond which Bragg's law is not satisfied. Practically, ϵ is about three times $\frac{1}{2} \Delta\vartheta_{\text{FWHM}}$.

The length l_c does not appear in [5] because along the analysing crystal, the reflecting region is limited by the scatterer length l_s and the limiting angles $\vartheta_{\text{Bragg}} - \epsilon$ and $\vartheta_{\text{Bragg}} + \epsilon$. Consequently, the scatterer must be large enough to exploit all of the analyser length.

Considering S approximately proportional to r (Fig. 6), we conclude from [5] that S is approximately inversely proportional to A_c .

Graphite is superior to other crystals (5) insofar as diffracting power is concerned. Consequently, graphite

is suitable for weak excitations, provided that the energy shift from the primary line is large. Such is the case for the X-ray Raman effect.

IV. Experimental results and discussion

The theoretical calculations of Sects. II and III were tested in a spectrometer constructed on the principles described above, for two scatterer-analyser distances, $r = 125$ and 209 cm. The analysing graphite crystal was provided by Union Carbide Corporation.

The following scatterers were used: chromium, colloidal graphite, amorphous boron, lithium, and polycrystalline graphite.

The scatterer was irradiated with a Cr X-ray tube ($U = 55$ kV, $i = 50$ mA), at a distance of 8 cm in two ways, transmission and reflection. A Geiger counter scanned the K_β primary line after reflection on the (0006) planes of the analyser.

The experimental results for the resolution S and for the intensity ratio $I(z)/I(0)$ are given in Table 1.

The theoretical values of the resolution for $r = 125$ and 209 cm deduced from Fig. 6 are $S_{125} = 105$ and $S_{209} = 164$, which are in adequate agreement with the

TABLE I. Experimental values of resolution S and the intensity ratio I_{125}/I_{209}

Scatterer	Scatterer-beam position	S_{125}	S_{209}	$\frac{I_{125}}{I_{209}}$
Chromium (polycryst.)	Transmission	152.3	201.5	2.05
	Reflection	130.4	201.5	2.03
Graphite (colloidal)	Transmission	134.4	184.7	1.38
	Reflection	147.8	177.4	1.62
Boron (amorphous)	Transmission	116.7	184.7	1.55
	Reflection	116.7	184.7	1.89
Lithium (polycryst.)	Transmission	100.8	130.4	1.93
	Reflection	113.6	184.7	2.16
Graphite (polycryst.)	Transmission	130.3	211.1	1.46
	Reflection	130.3	233.4	1.20
$\bar{S}_{125} = 127 \pm 5$ $\bar{S}_{209} = 189 \pm 8$ $\overline{I_{125}/I_{209}} = 1.7 \pm 0.1$				

corresponding experimental values $\bar{S}_{125} = 127$ and $\bar{S}_{209} = 189$.

The ratio I_{125}/I_{209} of peak intensities is exactly the ratio $A_{c_{125}}/A_{c_{209}}$. The experimental value $\overline{I_{125}/I_{209}} = 1.7$ is in good agreement with the theoretical value $209/125 = 1.67$ from [5].

The experimental ratio 1.7 of the peak intensities is close to the ratio $\bar{S}_{209}/\bar{S}_{125} = 1.5$ so we can accept the

theoretically deduced conclusion that I is approximately inversely proportional to S .

The constructed spectrometer is being used for X-ray inelastic scattering experiments with satisfactory results (6-8).

V. Acknowledgments

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1. L. V. AZAROFF. Elements of X-ray crystallography. McGraw Hill. New York. 1968. p. 552.
2. B. E. WARREN. X-ray diffraction. Addison Wesley, New York. 1969. p. 257.
3. UNION CARBIDE. Catalog Section EX-A-4110. 1970.
4. A. NILSON. Ark. Fys. **6**, 516, (1953).
5. R. W. GOULD and S. R. BATES. Appl. Spectros. **22**, 549 (1968).
6. D. K. LEVENTOURI, S. S. VAIPOULOS, A. B. VASSILIKOU, and C. N. KOUMELIS. Can. J. Phys. **56**, 438 (1978).
7. C. A. LONDOS and C. N. KOUMELIS. Solid State Commun. **31**, 735 (1979).
8. C. N. KOUMELIS and C. A. LONDOS. Can. J. Phys. **58**, 1507 (1980).