Plasmon Excitation in Compton-Scattering Experiments*

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X radiation (Cr $K\beta = 2.08$ Å) scattered by lithium, beryllium, and graphite at angles of 5°, 10°, and 15° was analyzed. Besides the known Rayleigh and Compton radiations, a new component, characteristic of the scattering substance, was observed and attributed to plasmon excitation. Curves giving the plasmon dispersion relation were drawn from which the values of the plasmon excitation energies for lithium, beryllium, and graphite were found to be 8.1, 20.7, and 6.5 eV, respectively. These results agree with the theoretical predictions of Nozieres and Pines (1959) and with previous experiments, such as fast-electron scattering (measurements of the characteristic energy loss) and optical measurements. For various reasons, such plasmon lines were not observed in many other experiments. In most cases the radiation used was too hard and the scattering angle too large; thus the conditions necessary for the appearance of the plasmons were not realized. However, there is one case (Theodossiou and Vosnidis, 1966) where the line might have been observed in lithium. A possible explanation for its nondetection is given.

I. INTRODUCTION

The spectrum of x rays diffusely scattered by solid materials has been the object of many investigations. Most of the earlier work[1] was restricted to relatively hard radiation, usually Mo radiation, and to relatively large scattering angles, usually 90° or larger. The scattered radiation was found to consist of two separate components: (a) a line due to the thermal diffuse scattering, and (b) a Compton band shifted towards lower energies. In the above work, DuMond and his co-workers succeeded in calculating the momentum distribution function of electrons, for the materials under examination by the Compton band. Recently, similar experiments have been reported.[2] On the other hand, Das Gupta[3] investigated a number of conductors, and found a radiation component, shifted from the primary line, corresponding to a loss of energy equal to the excitation energy of the inner electrons. Although this effect was not confirmed by Schulke and Berg[4] and by Weiss[5] it was observed by Faessler and Muhle[6] and by Suzuki.[7]

In some of the latest works,[8,9] a considerably smaller momentum transfer was involved, so that $k = (4\pi/\lambda)\sin\frac{1}{2}\varphi$ was in most cases smaller than the wave vector of the Fermi sphere. This was done for the purpose of investigating special properties of the valence electrons in the crystalline state. The first spectral investigation in the region of small momentum transfer was done in diamond[10] which is an insulator. It indicated that energy transfers to the valence electrons smaller than the energy gap are not possible. Similar measurements were extended to metallic Li[8,9] in the hope of determining the form of the Compton band, which, according to the one-electron theory, should consist of a straight line that continues into a parabola. Experimental errors, unfortunately, did not allow the verification of this form nor of the form calculated in the random-phase approximation[11] for single-electron excitation.

In 1959, Nozieres and Pines[12] calculated the spectrum of x radiation scattered by valence electrons. They found that it can be described by the imaginary part of the inverse dielectric constant $\mathrm{Im}\,\epsilon(k\omega)^{-1}$ exactly as in the case of electrons passing through matter. The spectrum consists of two components, the first corresponding to the usual Compton scattering and the second being due to the collective oscillation of the electrons (plasmons). The second component appears clearly only under the condition that k is smaller than a critical wave vector k_c given by

$$k_c = 0.47\sqrt{(r_s)}\,k_0 \quad , \tag{1}$$

where k_0 is the Fermi wave vector and r_s is the mean interelectron spacing measured in units of the Bohr radius. It was shown that the plasmon frequency ω_p follows the dispersion relation

$$\hbar\omega_k = \hbar\omega_p + (3\hbar\epsilon_F/5m\omega_p)k^2 \quad , \tag{2}$$

where

$$\omega_p = (4\pi ne^2/m)^{1/2} \quad , \tag{3}$$

ϵ_F is the Fermi energy, k the momentum transfer during the scattering, and n the electron density in configuration space. During an experimental investigation of the spectrum of x radiation scattered by lithium, beryllium, and graphite, a new component was detected by Priftis *et al.*[13] Further measurements have strengthened conviction that it

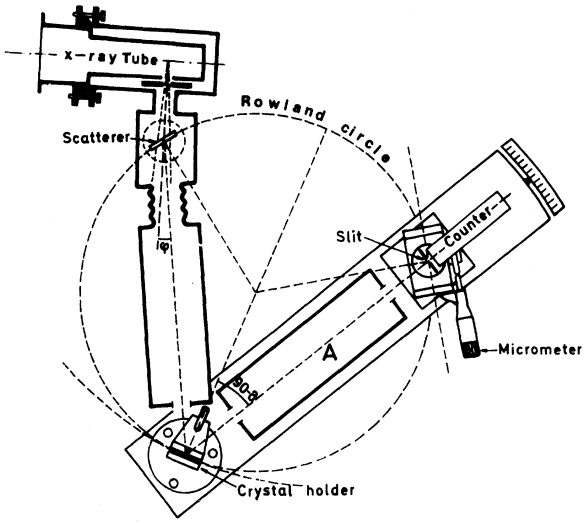

FIG. 1. Spectrometer, with the scatterer in the transmission position.

is due to the production of collective oscillations by the valence electrons. Moreover, in the most recent paper by Tanokua *et al.*,[14] evidence of plasmon excitation by x rays was reported. In Sec. IV, the reasons for which this plasmon component was not observed in earlier experiments are explained.

II. EXPERIMENTAL PROCEDURE

The experiments were carried out with the apparatus described by Theodossiou and Vosnidis[9] after the following modifications were made: The Cr x-ray tube was replaced by a larger one operating at 30 kV and 30 mA. The tube head is in a vacuum chamber in which the scatterers were positioned at a distance of 8 cm from the focus. Scattering takes place in a "transmission" position (Fig. 1). The spectrometer was adjusted to the spectral region near the $K\beta$ line.

A. Scattering By Metallic Lithium

The spectrum of the radiation scattered on Li at scattering angles $\varphi = 5°$, 10°, and 15° is shown in Fig. 2. The abscissa represents vernier reading of the micrometer along the Rowland circle. The energy increases towards the right. Under the above conditions the thermal diffuse scattering (TDS) and the Compton components overlap, and therefore knowledge of the primary spectrum, which is practically identical to the TDS, is required in order to separate them. This is done for each scattering angle in "dummy" experiments in which the scatterer is replaced by a sample that has the same dimensions as the lithium scatterer and consists of paraffin and Cr_2O_3 powder. The hard part of the radiation emitted by the tube makes

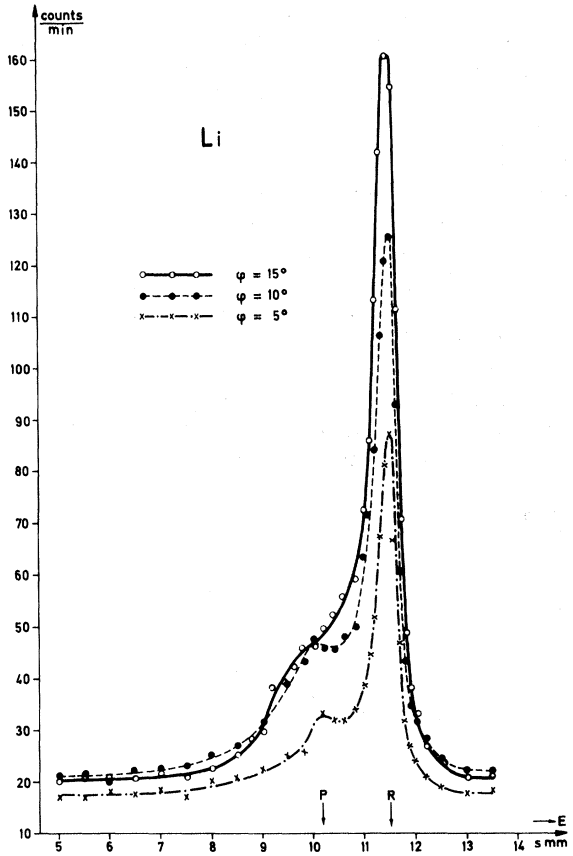

FIG. 2. Spectrum of the radiation scattered by Li at scattering angles $\varphi = 5°$, 10°, and 15°. On the abcissa s represents vernier readings of the micrometer along the Rowland circle. The energy (E) increases towards the right. The arrows R and P indicate the position of the TDS and the plasmon component, respectively.

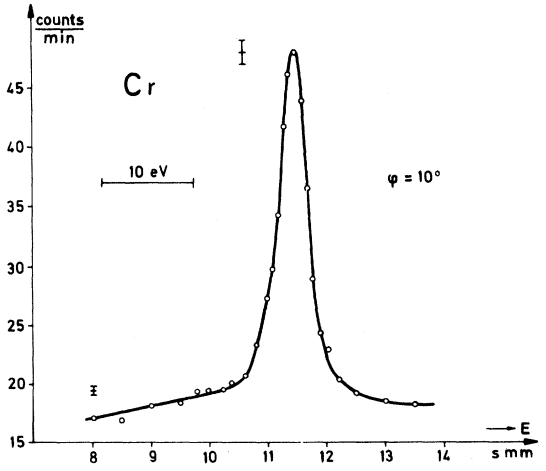

FIG. 3. Primary radiation spectrum (dummy experiment) at $\varphi = 10°$. On the abscissa, s represents vernier readings of the micrometer along the Rowland circle. The energy (E) increases towards the right.

the chromium atoms fluoresce. This radiation (Fig. 3) is free from any Compton component and hence corresponds exactly to the primary spectrum. Owing to the geometrical similarity of the sources of both radiations, i.e., the lithium scatterer in the main experiment and the fluorescing chromium in the dummy experiments, any distortion of the spectrum owing to the astigmatism of the apparatus becomes ineffective. By adjusting[8,9] the curve of Fig. 3 to Fig. 2 and taking the difference, we obtain Fig. 4. This gives the spectrum of the scattered radiation after elimination of the TDS and corresponds to $\mathrm{Im}\,\epsilon(k\omega)^{-1}$. As it is explained in the next paragraph, the "hump" on the left side is due to plasma excitation, while the band on the right side is the Compton band.

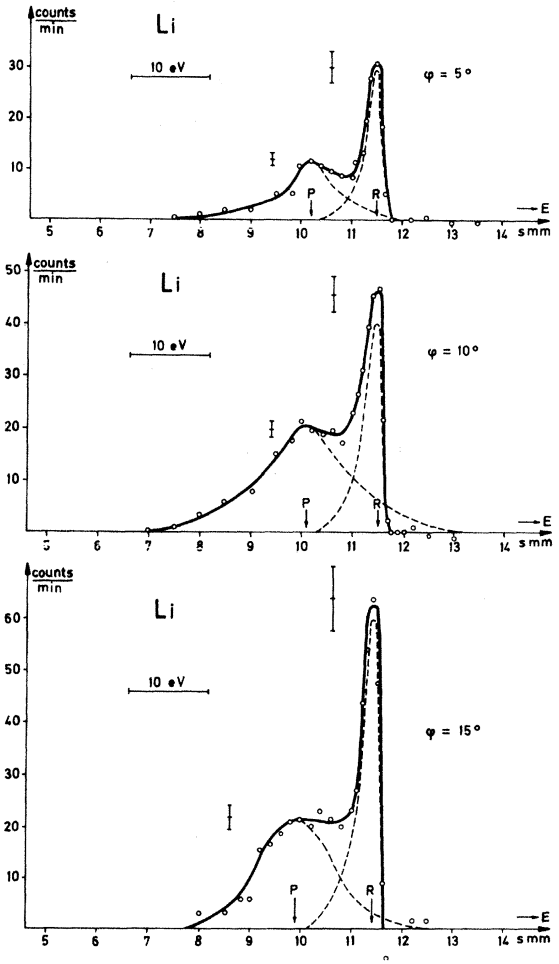

FIG. 4. Spectrum of the incoherently scattered radiation by Li at scattering angles $\varphi = 5°$, 10°, and 15°,. On the abscissa, s represents vernier readings of the micrometer along the Rowland circle. The energy (E) increases towards the right. R: position of the peak of the primary radiation. C: position of the classical Compton shift for stationary electrons. P: position of the peak of the plasmon line.

The dashed lines in Fig. 4 show the separation of the one-electron Compton profile from the plasmon band. The technique used for this separation was as follows: We find the zero position of the energy loss from the top of the primary radiation (position R in Fig. 4). Now, since it is known that the Compton band is extended as far from R as $\Delta\lambda + \delta\lambda$, where $\Delta\lambda = (h/mc)(1 - \cos\varphi)$ is the classical Compton shift, and $\delta\lambda = 2(v_F/c)\lambda_0 \sin\frac{1}{2}\varphi$ (v_F is the velocity at the top of the Fermi distribution, c is the velocity of light, λ_0 is the wavelenghth of the incident radiation, and φ the scattering angle),[15] we find by simple calculations for all these scattering angles for Li and Be that the energy corresponding to the $\Delta\lambda + \delta\lambda$ is smaller than that of the plasmon band top. This, in fact, means that the Compton band does not affect the plasmon band top at all.

On the other hand, according to the theory, a plasmon is described by a δ function; so we can assume that the observed plasmon band, which is wider, because of the not perfectly monochromatic primary radiation and the effect of the spectrometer, will have the shape of the primary radiation which is symmetrical to a good approximation (Fig. 3). Thus we can draw first the whole plasmon-band profile thanks to the symmetry, and then, the Compton profile just by subtraction of the former from the initially obtained curve (continous line in Fig. 4).

Comparing the three Compton bands in Fig. 4, we observe that, in agreement with the theory, there is an increase of the intensity and of the area of the Compton band with increasing scattering angle. The Compton profile, however, seems to disagree with that observation, which was predicted by any of the theories (see Ohmura and Matsudaira's[11] Fig. 3). An exact statement on the form that would result from the our experiments cannot be made without deconvoluting the experimental curve.

Now, we will discuss the band on the left-hand side of Fig. 4. During the first experiment, it was assumed that the hump in question was due to the "astigmatic errors" of the apparatus or to some parasitic radiation. These assumptions were soon rejected, for the following reasons: (i) After shifting the scatterer to various positions, the same hump appeared. (ii) Experiments conducted after removing the scatterer gave a horizontal background. (iii) The "dummy" experiments, although subjected to the same astigmatic errors, gave no "hump."

B. Scattering by Beryllium and Graphite

The results are shown in Figs. 5 and 6. The TDS and the Compton components overlap, and no effort was made to separate them. Towards the

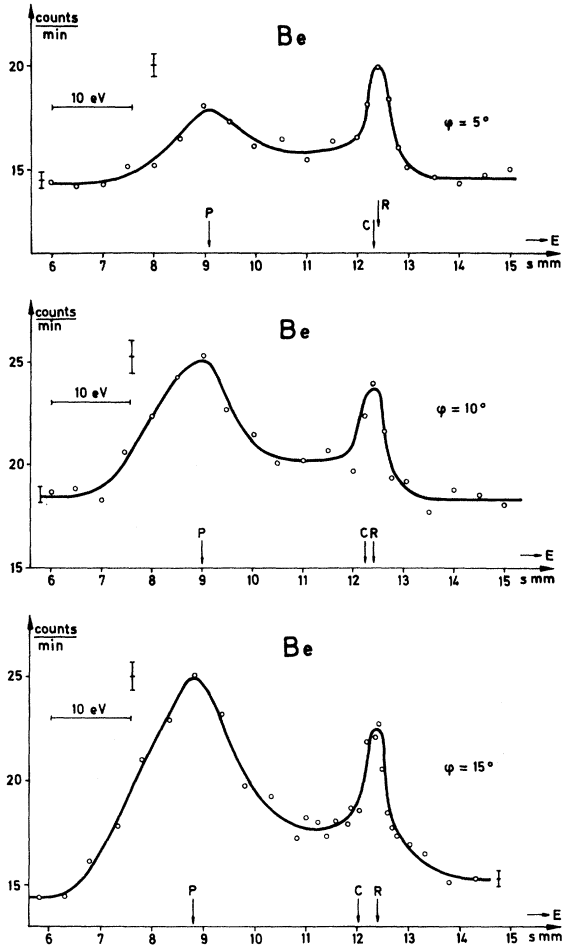

FIG. 5. Spectral analysis of the scattered radiation by Be. For detail see caption of Fig. 4.

left, a hump marked P appears in all cases. It was noticed that this hump appears at a different distance from the initial line marked R for each material.

III. PLASMONS

The above results suggest that the position of the hump depends on the substance; this is equivalent to stating that there is a different energy loss for each substance used as the scatterer. As the observed energy losses for lithium, beryllium, and graphite coincide with the plasma excitation energy values determined by other methods, we ascribed the humps to plasmons. The energy shift of the plasmon line according to Eq. (2) depends on the scattering angle (i.e., on k). The experimental shifts for Li and Be are plotted in Fig. 7 (crosses) as a function of k^2. The estimated errors are indicated with vertical and horizontal lines. They are large because the position of the maximum of the plasmon line cannot be determined with accuracy, and because the scattering angle varies up to 1.5°, for radiation originating in the various points of the scatterer. By extrapolating the results to $k=0$, we obtain an energy value which we define as the plasmon energy. The results for the three materials examined are shown in Table I. In the same table are the plasmon energies determined either from the characteristic energy loss of electrons in thin films or from the optical constants. The values listed under "theory" have been calculated by Pines[16] from formula (3) for a free-electron gas. The value of $\hbar\omega_k$ is plotted in Fig. 7 according to Eq. (2), by using the theoretical values for $\hbar\omega_p$ and the values of 4.8 and 14.6 eV for the Fermi energy of Li and Be.

The half-width of the plasmon lines which, ac-

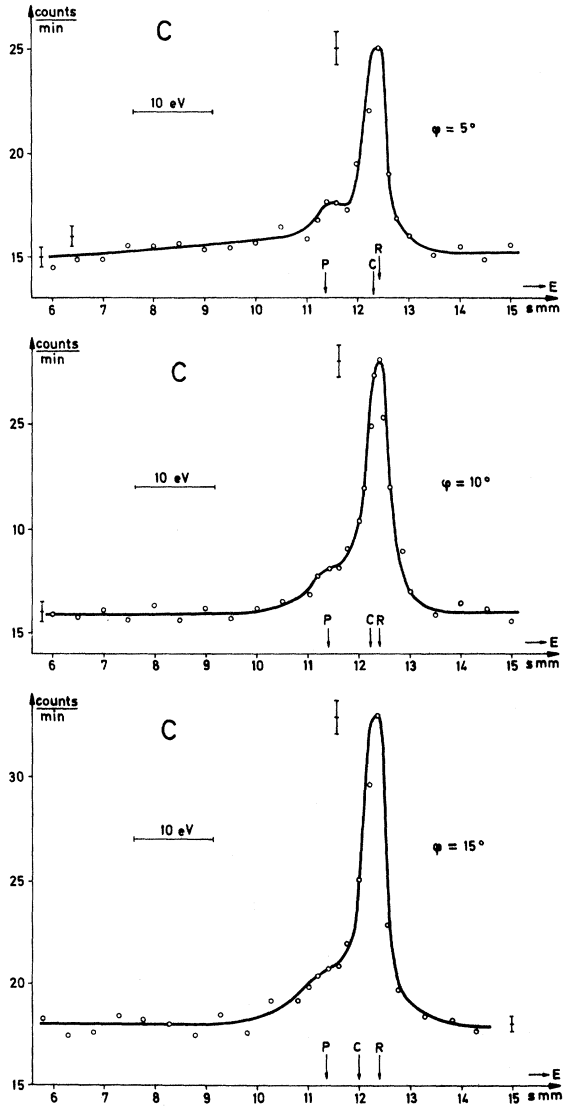

FIG. 6. Spectral analysis of the scattered radiation by graphite. For details see caption of Fig. 4.

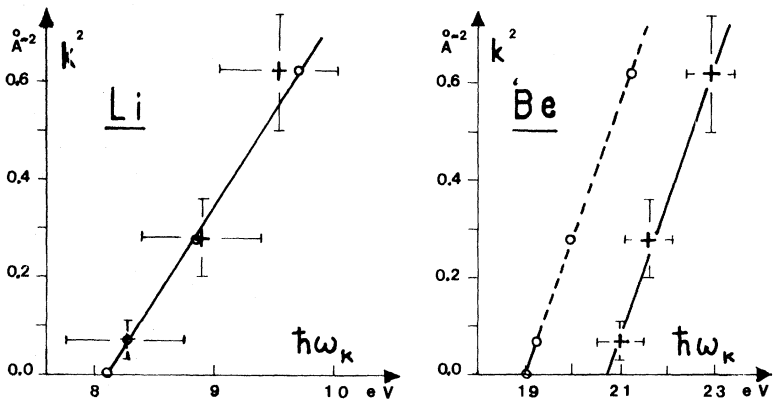

FIG. 7. Plasmon dispersion relation for Li and Be. The energy of the plasmon line $\hbar\omega_k$ is plotted as a function of k (momentum transfer). The experimental values are indicated by crosses. The theoretical values, indicated by circles, are calculated according to formula (2).

cording to the theory, depends on the plasmon lifetime, was found to be about three times wider than that found in fast-electron scattering experiments. This large width is partly due to the natural width of the primary radiation and to the widening caused by the apparatus itself. In this way, a width of approximately 3.2 eV is obtain, experimentally, for the primary radiation (Fig. 3). Although, as mentioned, the width of the plasmon line cannot be determined accurately without deconvoluting the experimental curve, the comparison of the curves, for various angles φ clearly shows that the greater the scattering angle φ, the wider the plasmon width, or, in other words, the damping of the plasmon increases with k, in complete agreement with the theoretical predictions.

TABLE I. Plasmon energy in eV.

Elements	Theory[a]	Other experiments	Present work
		8.0[b]	
Lithium	8.0	9.8[c]	8.1
		8.2[d]	
		18.9[e]	
		19.1[f]	
Beryllium	19.0	19.0[g]	20.7
		19.9[d]	
		18.9[h]	
		7.3[g]	
Graphite	7.0[i] 25.0	7.2[j] 25.1[j]	6.5
		7.5[k] 26.0[k]	
		7.0[l] 25.0[l]	

[a]As caluclated by D. Pines in Ref. 12.
[b]R. W. Wood and C. Lukens, Phys. Rev. 54, 332 (1938).
[c]L. Marton, L. Leder, and H. Mendlowitz, in *Advances in Electronics and Electron Physics*, edited by L. Marton (Academic, New York, 1955), Vol. 7.
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TABLE II. List of k's involved (in Å^{-1}) in Compton experiments by different authors. The values of k_c for Li and Be are 0.953 and 1.25, respectively.

Authors	Li	Be	Graphite
DuMond (1933)		17.6	$\geq$ 9.35
Kappeler (1936)	$\geq$ 4.64		$\geq$ 4.64
Kirkpatrick and Dumond (1938)			16.5
Alexopoulos and Brogren (1953)		1.21	
		1.45	
		1.80	
Das Gupta (1962)	3.12	$\geq$ 2.79	
Cooper *et al.* (1965)[a]	$\simeq$ 12.5	$\simeq$ 12.5	
Alexandropoulos and K. Alexopoulos	$\simeq$ 1.4		
(1965) Theodossiou and Vosnidis (1966)	0.526		
	0.945		
	1.67		
	2.31		
Faessler and Muhle (1966)		4.37	
Cooper and Leake (1966)[b]			$\simeq$ 14.5
Schulke and Berg (1967)		4.53	
Suzuki (1967)	$\geq$ 5.2	$\geq$ 2.11	$\geq$ 2.11
Phillips and Weiss (1968)	$\simeq$ 14.9	$\simeq$ 14.9	$\simeq$ 14.9
Present work	0.263	0.263	0.263
	0.526	0.526	0.526
	0.789	0.789	0.789

[a]M. Cooper, J. A. Leake, and R. J. Weiss, Phil. Mag. 12, 797 (1965).
[b]M. Cooper and J. A. Leake, Phil. Mag. 18, 603 (1966).

IV. COMPARISON WITH EARLIER EXPERIMENTS

Plasmon excitation was never observed in any of the earlier measurements of the Compton spectrum. The main reason for this is considered to be the excessively high value of the momentum transfer, i.e., $k > k_c$. In Table II, the values of k are listed.

In some experiments,[8,9,10] k was found approximately equal to k_c. In such cases, the theory is only partially valid. The momentum transfer results in an intermediate state between a collective electron behavior and a single-particle behavior, and the plasmon is damped and does not represent a well-defined excitation. In addition, the plasmon line is masked by the Compton band which is shifted and widened in such a way that it covers the plasmon band.

In the measurements made on lithium by Theodossiou and Vosnides,[9] there is a case ($\varphi = 10°$) in which conditions were similar to ours. However, no plasmon line was observed. This is probably due to the larger power of the x-ray tube which was used in our experiments. It also might be due to the fact that in the "reflexion" position of the scatterer, the slightest change of the position of the scatterer produced a different spectrum, which was not the case when the scatterer was in the "transmission" position.

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[15]Obviously the 1_s core electrons are too tightly bound to contribute to the scattering.

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