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Inelastic photon scattering from *K*-shell electrons of Cu and Zr

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The inelastic scattering cross section from the tightly bound 1*s* electrons at the intermediate momentum transfer from Cu and Zr was measured using a 59.54-keV γ-ray source and a coincidence technique. A good statistical accuracy was achieved by using a NaI-scintillation counter for the fluorescence detection. The experimental results are compared with the impulse approximation which seems to explain the spectral shape surprisingly well. Neither the Compton peak shift nor any predicted structure in the measured spectra is observed.

During the last few years considerable interest has been directed to the inelastic photon scattering from inner-shell electrons. Especially the accuracy and the possible failure of the impulse approximation has been studied by varying the energy and momentum transfer in the scattering process. Because the inner electron shell in most cases has only minor contribution to the total inelastic scattering cross section, a coincidence technique utilizing simultaneous x-ray fluorescence has been generally used in these studies.

Namikawa and Hosoya[1] (NH) used 60-keV γ rays to study inelastic scattering by the Cu *K*-shell electrons at the momentum-transfer k close to the value which according to the theory would lead to the Raman-type transitions in atomic systems[2] and in metals[3] related to the breakdown of the impulse approximation. This interesting region can be described by $ka \approx 1$, where a is the Bohr radius of the desired electron orbit. Close to the absorption edge NH found a Raman-type peak and an additional contribution explained to be due to the double Thomson scattering. They also found a shift in the Compton peak due to the electron binding. It was later supposed by Manninen[4] that the observed structure was largely due to the false coincidence events. This was supported by the calculations on double Thomson scattering by Marchetti and Franck[5] (MF) and Ohmura and Satō[6] (OS) as well as the experiments by MF (Ref. 7) and Manninen *et al.*[8]

The aim of the present work is to improve the experimental accuracy and simultaneously measure the *K*-shell distribution of a heavier material in order to check the validity of the theory. For that purpose a large area scintillation counter is used to detect x-ray fluorescence. In order to compare the new experimental system to the previous ones a Cu target has been used. Measurements on Zr having the *K* binding energy of 18 keV compared with 9 keV of Cu gives a more critical test to the impulse approximation.

Measurements were accomplished using a 900-mCi ^{241}Am source that emits 59.537-keV γ-rays. A lead tube with an inner silver hole to prevent Pb*L*-fluorescence radiation from entering the x-ray detector was placed between the source and the sample to collimate the incident γ rays. Inelastically scattered photons from the sample foils were measured using an intrinsic Ge detector (thickness 5 mm, diameter 10 mm) and *K*-fluorescence radiation was detected using a NaI-scintillation counter which has a diameter of 2 in. The total efficiency, including the geometrical factor of the NaI detector was high (8%) while the energy resolution (2.5 keV at 8 keV) was quite poor: The fluorescence energy window including overlapping $K\alpha$ and $K\beta$ contributions in the case of Cu was 4.8–11.5 keV. The source, the sample foil, and the Ge detector were on the same horizontal plane, the scattering angles being 125° and 128° for Cu and Zr, respectively. The NaI detector was placed above the Ge detector at the angle of 60° relative to the plane. The source-sample distance was 13 cm and the sample-detector distance was 4 cm for both detectors. The sample foil face was towards the fluorescence detector in order to minimize the fluorescence x-ray path in the sample and to increase the irradiated volume. The samples were polycrystalline foils with thicknesses of 17 μm (Cu) and 30 μm (Zr). A relatively large angular divergence ($\pm 12°$) for the scattered γ rays was accepted to improve the low counting rate.

Timing information for the coincidence electronics was taken out from the special fast timing output of the Ge-detector's preamplifier and from the photomultiplier anode of the NaI detector. A fast-slow coincidence circuit with a constant fraction timing mode was used to gate a multichannel analyzer measuring inelastically scattered photons. The time resolution measured with a time analyzer appeared to be better than 20 ns full width at half maximum (FWHM), but a 35 ns time window was used in the measurements in order to include all true coincidence events.

A 1024-channel multichannel analyzer was used in the measurements but to improve the counting statistics the contents of the 16 adjacent channels were added together, which gives a channel width of about 1 keV. The measurements were carried out in periods of about a day to check the stability of the system and the total measuring time was 240000 s for the Cu and 675000 s for the Zr sample. The total counting rate in the coincidence mode in the energy range of 20–65 keV was 0.044 cps for the Cu and 0.055 cps for the Zr sample. In order to subtract the chance coincidence events the signal from the Ge detector was delayed in the Cu measurement by 150 ns. The measured true-to-total coincidence-counting-rate ratio was 75% compared with 47% in the previous γ-ray experiment[1] and about 5% in the synchrotron experiment.[7]

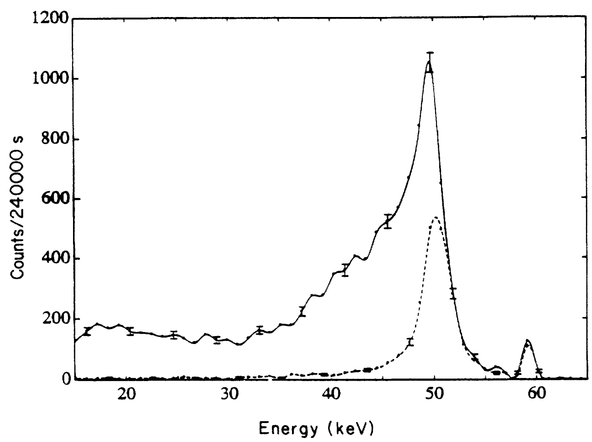

FIG. 1. Total (solid line) and chance (dashed line) coincidence spectrum of Cu. Continuous line is spline fitting to the experimental data points drawn with statistical error bars.

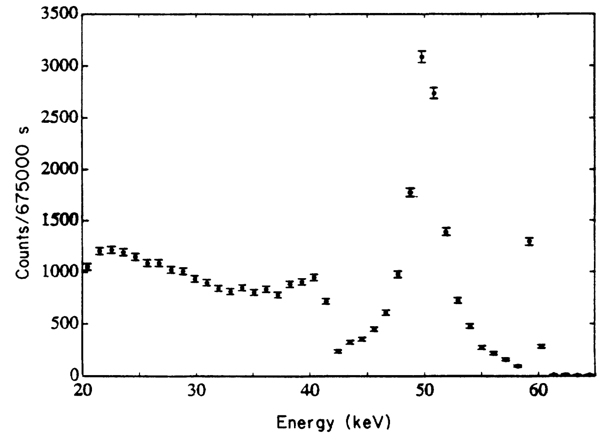

FIG. 2. Total experimental coincidence spectrum of Zr. Vertical line shows the K-edge position.

Figure 1 shows the measured coincidence and the chance coincidence spectrum for Cu. It can be seen that most chance coincidence events are in the energy range above the K edge at 50.56 keV which is close to the center of the energy spectrum produced by outer electrons (at 50.3 keV) and thus their subtraction lowers the statistical accuracy only around the absorption edge. In the case of Zr (Fig. 2) the K absorption edge at 41.54 keV is well below the central peak due to the outer-shell electrons and the effect of the chance events in the interesting energy range is almost insignificant. In this case the chance coincidence contribution was approximated by measuring the energy spectrum without the coincidence gating and equalizing the spectrum area above the K edge to the measured coincidence spectrum in this area. This method turned out to be good enough which can be seen from the absence of the elastic and outer-shell contribution in the final processed spectrum in Fig. 4.

It can clearly be seen in Figs. 1 and 2 that the measured spectra contain, in addition to the K-shell Compton profile, some other low-energy contribution. This feature is even more pronounced after the absorption correction. Besides the low-energy noise in the electronic circuit and the escape effects in the Ge detector, which do not give noticeable contribution in our energy range of interest (20–60 keV), there are two main possibilities.

(a) Infrared divergence of the scattering cross section.[9] This is related to the $\mathbf{p}\cdot\mathbf{A}$ term in the interaction Hamiltonian and its effect in similar conditions has been recently estimated.[10] In our case the inelastic scattering due to the $\mathbf{A}^2$ term dominates and at the present level of experimental accuracy we can neglect the $\mathbf{p}\cdot\mathbf{A}$ contribution.

(b) Bremsstrahlung due to the K-shell photoelectrons. For the Cu target and 60-keV primary radiation, photoelectric absorption has a major role in the interaction between the photons and the target electrons and its contribution to the total absorption cross section is about 85% and is mainly due to the K electrons. Photoelectrons released in the absorption process slow down in the target material emitting radiation. In the case of K-shell photoelectrons it is followed by K fluorescence and a true coincidence signal can be detected. As pointed out by MF (Ref. 10) this is a significant component in our desired energy range and should be subtracted before reliable information about inelastically scattered photon spectrum can be extracted.

The amount of bremsstrahlung depends on the thickness of the target. For "thick" samples the differential cross section can be written[10,11]

$$\frac{d^2\sigma}{d\Omega\, d\omega_2}=C\frac{T-\hbar\omega_2}{\hbar\omega_2}\,, \tag{1}$$

where C is a proportional constant, T the kinetic energy of the photoelectron, and $\hbar\omega_2$ the energy of the radiated photon. The target can be considered infinitely thick as soon as it is thicker than the mean free path of the photoelectron, which is in our case certainly true.

Raw data shown in Figs. 1 and 2 were processed in the following way. The chance coincidence contribution was first subtracted as described above and the spectra were corrected for the sample absorption. Because our cross-section measurement was not an absolute one the spectral line shape of bremsstrahlung, given by Eq. (1) was fitted to the absorption corrected experimental data in the energies which were low enough to include any noticeable Compton scattering contribution and by demanding the bremsstrahlung contribution to be zero above the K edge. In Fig. 3 the spectrum division into the bremsstrahlung and the inelastic scattering part can be seen in the case of Cu.

The comparison between the experimental results and the theory was done in terms of the differential scattering cross section. The theoretical 1s-electron Compton profiles for Cu and Zr were taken from Biggs, Mendelsohn, and Mann.[12] The theoretical values given in atomic units of electron momentum were converted to the energy scale using the well-known relation between the differential scattering cross section and the Compton profile.[13] It should be noted that often referred formulas[14] for this transformation given erroneous results at the high momentum and therefore the more exact result[13] should

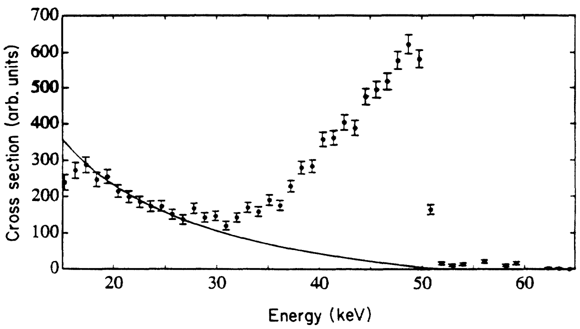

FIG. 3. Coincidence spectrum of Cu after the subtraction of chance coincidences and the correction for sample absorption. The solid line is the bremsstrahlung contribution based on Eq. (1). The decrease of the measured intensity below 18 keV is due to the threshold discriminator. The vertical line shows the K-edge position.

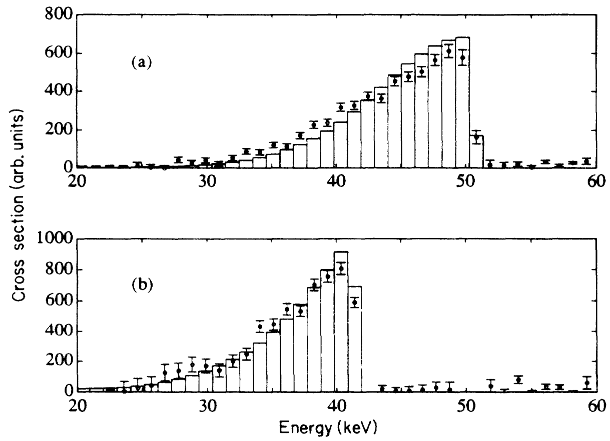

FIG. 4. Final experimental inelastic scattering cross section (♦) for K electrons of (a) Cu and (b) Zr. The theoretical cross sections (solid bars) are based on Compton profiles, calculated using impulse approximation.

be used.

To include the binding-energy cutoff the theoretical cross section was truncated at the energy corresponding to the K binding edge. The interpolation procedure leading to the same channel width as in the experiment followed by the adding of 16 channels allowed the comparison between the theory and the experiment. The resolution smearing of the theoretical spectrum at the binding edge was then mainly due to the addition of channels, the effect of the detector resolution (400 eV FWHM at 60 keV) was a much smaller factor.

Final results are shown in Figs. 4(a) and 4(b). The theoretical values of $d\sigma/d\Omega$ were normalized according to the experimental ones by integrating in the energy range from 20 keV to the energy corresponding to the binding edge. One can immediately see that the theoretical model based on the impulse approximation works surprisingly well although we now have $ka = 0.94$ for Cu and 0.67 for Zr, calculated at the absorption edge. Compared with the previous experiments and calculations we can also conclude the following:

(i) Within the present resolution there is neither the Raman-type peak close to the absorption edge, predicted by OS (Ref. 3) nor the double Thomson peak observed by NH.[1] With a better resolution it is, of course, possible to find x-ray absorption near-edge structure and extended x-ray absorption fine-structure oscillations but it is out of the scope of the present work.

(ii) Although we are now at the momentum-transfer region where the impulse approximation should not work and one would therefore expect to see the effect of the binding energy in the Compton shift,[1,3] there is no evidence for such shift.

(iii) The present results on Cu agree well with those obtained using synchrotron radiation.[7] MF used somewhat higher incident photon energy (70 keV) and due to the unfavorable change-to-true coincidence ratio the experimental error close to the absorption edge was significantly larger than in our case. Additionally the present result for Zr, which is an even more significant test due to the smaller ka value, shows that within the experimental uncertainty the impulse approximation still works.

A well-collimated characteristic x-ray beam from a high-voltage x-ray tube opens new possibilities in the inner-shell coincidence studies. It is then possible to measure both $d^2\sigma/d\Omega d\omega_2$ and $d\sigma/d\Omega$ in absolute scale as a function of momentum transfer. On the basis of such studies, which are in progress in our laboratory, more information about the inelastic scattering cross section can be obtained.

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[1]K. Namikawa and S. Hosoya, Phys. Rev. Lett. **53**, 1606 (1984).
[2]P. Eisenberger and P. M. Platzman, Phys. Rev. A **2**, 415 (1970).
[3]Y. Ohmura and S. Satō, J. Phys. Soc. Jpn. **56**, 1657 (1987).
[4]S. Manninen, Phys. Rev. Lett. **57**, 1500 (1986).
[5]V. Marchetti and C. Franck, Phys. Rev. A **35**, 3128 (1987).
[6]Y. Ohmura and S. Satō, J. Phys. Soc. Jpn. **57**, 3599 (1988).
[7]V. Marchetti and C. Franck, Phys. Rev. Lett. **59**, 1557 (1987).
[8]S. Manninen, K. Hämäläinen, T. Paakkari, and P. Suortti, J. Phys. (Paris) Colloq. **48**, C9-823 (1987).
[9]Y. Bannett, D. Rapaport, and I. Freund, Phys. Rev. A **16**, 2011 (1977).
[10]V. Marchetti and C. Franck, Phys. Rev. A **39**, 647 (1989).
[11]R. D. Evans, *The Atomic Nucleus* (McGraw-Hill, New York, 1955).
[12]F. Biggs, L. B. Mendelsohn, and J. B. Mann, At. Data Nucl. Data Tables **16**, 201 (1975).
[13]R. Ribberfors, Phys. Rev. B **12**, 2067 (1975).
[14]M. J. Cooper, Rep. Prog. Phys. **48**, 415 (1985).