Observation of a New Component in the X-Ray Scattered Spectrum

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The results of a spectral analysis of Cu $K\alpha_1$, $K\alpha_2$ x-rays scattered by a single crystal of LiF (lithium fluoride) and by LiF powder (at scattering angles 16°, 25°, 70°, 84°) are evidence of the existence of a new line on the long-wavelength side of the spectrum. This line is 5 eV from the incident beam, and its position and shape are independent of the scattering angle. The peak intensity of the new line is of the same order of magnitude as the Compton intensity. The observed line can be considered to be caused by an "x-ray Raman"-type process from F-centers which themselves were produced by the incident x-ray beam. This is not the only possible explanation. However, the position of the line is comparable to the F-center absorption energy for LiF as measured by an ultraviolet spectrometer. The relative peak intensity indicates that the differential cross section for the process presently under investigation is higher than the Compton cross section for valence electrons.

INTRODUCTION

SINCE the earliest days of x-ray spectroscopy, the shape of the x-ray Compton band for large scattering angles has been used for studying the electron-momentum distribution in solids. In most cases, the impulse approximation was used. Here the energy transferred to the electron is very large compared to the electron binding energy, and the electron is ejected from the system before the system has had time to readjust. Therefore, the Compton differential cross section is a function of the unperturbed electron wave functions. However, some fine structure[1] and discontinuities on Compton bands caused by bound electrons have been observed. The differential cross section for the Compton scattering from a bound electron has been approximately calculated by Platzman and Tzoar[2] and is given by

$$\left(\frac{d\sigma}{d\omega\, d\Omega}\right)=\left(\frac{d\sigma}{d\Omega}\right)_{\rm Th}\sum_q |\langle q|e^{i\mathbf{k}\cdot\mathbf{x}}|\phi\rangle|^2\delta(\epsilon_q+E_B-\omega).$$

Here $(d\sigma/d\Omega)_{\rm Th}$ is the single-electron Thomson cross section, $|q\rangle$ and ϵ_q are the wave function and the energy, respectively, of the recoiling electron, $|\phi\rangle$ is the ground-state wave function of the bound electron, and $\mathbf{k}$ is the momentum transfer to system in the scattering process. According to this relation, the Compton spectrum is expected to consist of a series of lines and a continuous band. (Line and band correspond, respectively, to a discrete or free-electron state.) This prediction came first from Sommerfeld.[3] Discontinuities and lines characteristic of the scattering atoms have been observed. The application of the previous formula can be extended to other systems with discrete energy levels such as F centers. In this case, because of hydrogenlike energy structure, the incoherent scattering cross section is expected to be high.

EXPERIMENTAL RESULTS

Copper $K\alpha_1$, $K\alpha_2$ x rays were scattered by a single LiF crystal and by LiF powder at four scattering angles: 16°, 25°, 70°, and 84°. The scattered radiation was analyzed using a double crystal spectrometer[1] which provided a resolution of $E/\Delta E=3000$. Figure 1 is the spectrum near Cu $K\alpha_1$, $K\alpha_2$ of the scattered radiation from a single LiF crystal at a scattering angle of 84°. Spectra obtained using any of the four scattering angles (LiF single crystal or powder as scatterer) are similar to this one. Both lines are asymmetrical on the longer-wavelength side, the asymmetries starting at 5 eV from the peaks. Each of these asymmetrical lines can be considered to be the superposition of three spectral components: (a) a coherent component similar to the Cu emission line (symmetrical for all practical purposes), (b) an incoherent component with asymmetrical line shape and with a peak at 5 eV from (a), and (c) the Compton band starting at 10 eV from the coherent line.

The spectrum of the scattered radiation was analyzed into its components by comparison with the spectrum of Cu fluorescence,[4,5] Fig. 2. Figure 3 is a plot of the ratio: $(I_{\rm LiF}-{\rm BG}_1)/(I_{\rm Cu}-{\rm BG}_2)$, where $I_{\rm LiF}$ is the intensity of the scattered spectrum from LiF, ${\rm BG}_1$ is the background for the LiF measurement, $I_{\rm Cu}$ is the intensity of the Cu fluorescence, and ${\rm BG}_2$ is the corresponding background. This ratio is a linear function of

[1] N. G. Alexandropoulos and G. G. Cohen, Phys. Rev. **187** 455 (1969).
[2] P. M. Platzman and N. Tzoar, Phys. Rev. **139**, A410 (1965).
[3] A. Sommerfeld, Phys. Rev. **50**, 38 (1936).
[4] N. G. Alexandropoulos, Phys. Rev. **150**, 610 (1966).
[5] The Cu fluorescence spectrum was chosen as a reference because this is the only spectrum that is free of the Compton effect and still includes all the instrumental distortions. The spectrum of the fluorescence radiation is a better reference than the tube spectrum because it is taken under exactly the same conditions as the spectrum of the scattered radiation.

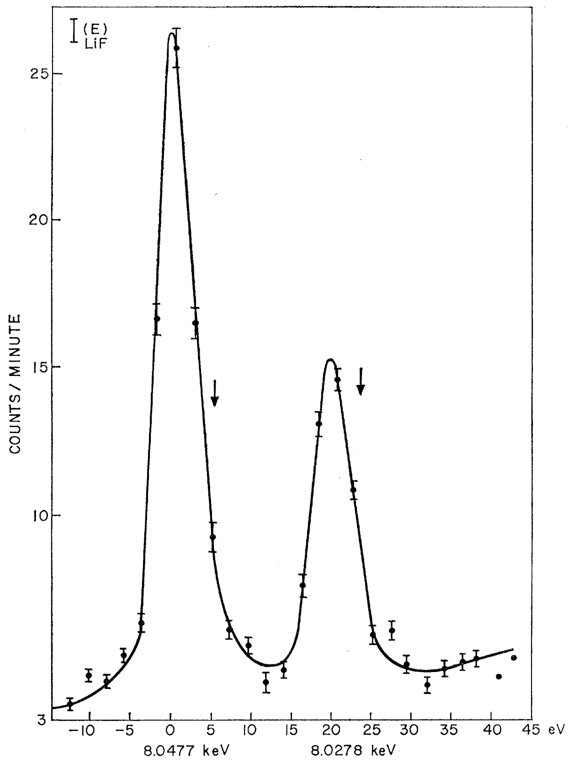

FIG. 1. Spectrum of Cu $K\alpha_1$, $K\alpha_2$ scattered by LiF at scattering angle 84°. The arrows indicate the discontinuities.

the differential cross section of the incoherent intensity. In Fig. 3, line A is the new incoherent component of the Cu $K\alpha_1$ scattered radiation; line B is the new component of the Cu $K\alpha_2$ superimposed on Cu $K\alpha_1$ Compton band; band C is the superposition of the Cu $K\alpha_1$, $K\alpha_2$ Compton bands. The position and the shape of lines A and B are found to be independent of the scattering angle; only their relative intensity, as compared with band C, depends on angle.

Although several processes could give rise to these lines, they most probably are x-ray Raman-type lines caused by transitions of the electrons of the *F* centers in LiF. These *F* centers are produced by the continuous and characteristic radiation of the incident beam; the estimated concentration of *F* centers is between 10^{15} to 10^{17} *F* centers per cm^3. A comparison of the concentration of *F* centers with the concentration of valence electrons indicates that the cross section of the process under investigation is higher than the Compton cross section. The precise relative magnitude of these components could be obtained from the ratio of the integrated intensities of the two spectral components.

Some fine structure has been observed in the incoherent part of the scattered radiation, i.e., the so-called "x-ray Raman lines,"[1,6,7] the plasma excitation line,[8] and discontinuities on the shorter-wavelength side of the Compton band due to binding effects.[9,10] The new observed line is not included in any of the above fine structure. At the same time, the existence of impurities in the sample or in the tube cannot account for the observed line.

The present results established[11] the existence of a new component in the scattered radiation but are inconclusive with respect to the shape of the observed line and the process which gives rise to it. The reported investigation is being continued using an x-ray experimental arrangement plus an ultraviolet spectrometer

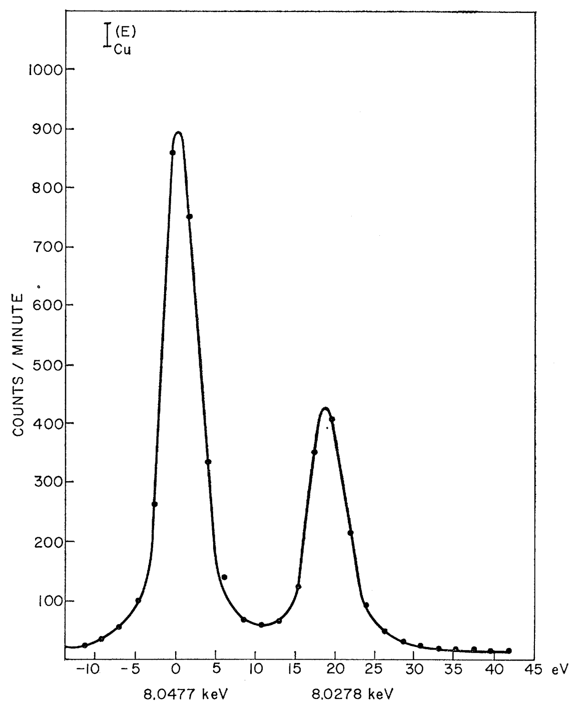

FIG. 2. Copper fluorescence spectrum obtained under exactly similar experimental conditions as in Fig. 1.

[6] K. Das Gupta, Phys. Rev. Letters **3**, 38 (1959).

[7] T. Suzuki, J. Phys. Soc. Japan **22**, 134 (1967).

[8] G. Priftis, A. Theodossiou, and K. Alexopoulos, Phys. Letters **27A**, 577 (1968).

[9] N. G. Alexandropoulos and K. Alexopoulos, Phys. Rev. **140**, A597 (1965).

[10] M. Cooper and J. A. Leake, Phil. Mag. **17**, 241 (1966).

[11] Each point in Fig. 1 is the average of three points, 0.72 eV apart, and each representing a minimum accumulation of 300 counts. The error in the Cu fluorescence spectrum is much smaller and can be considered to be negligible because of the high intensity. There is an ambiguity in the determination of BG_1 and BG_2 but the limits of the background values are very well defined. The variation in BG_1 and BG_2 within these limits does not change the general shape of Fig. 3 and the relative position of the peaks are reproducible to better than 10% for all checked background and for all eight different spectra (four for the single LiF crystal and four for the LiF powder). The estimated statistical error is 10% for the ratio, but in Fig. 3 a maximum error has been shown for several points, i.e., the ratio has been calculated for the improbable cases in which the BG_1 is the lower limit and BG_2 is the upper and vice versa.

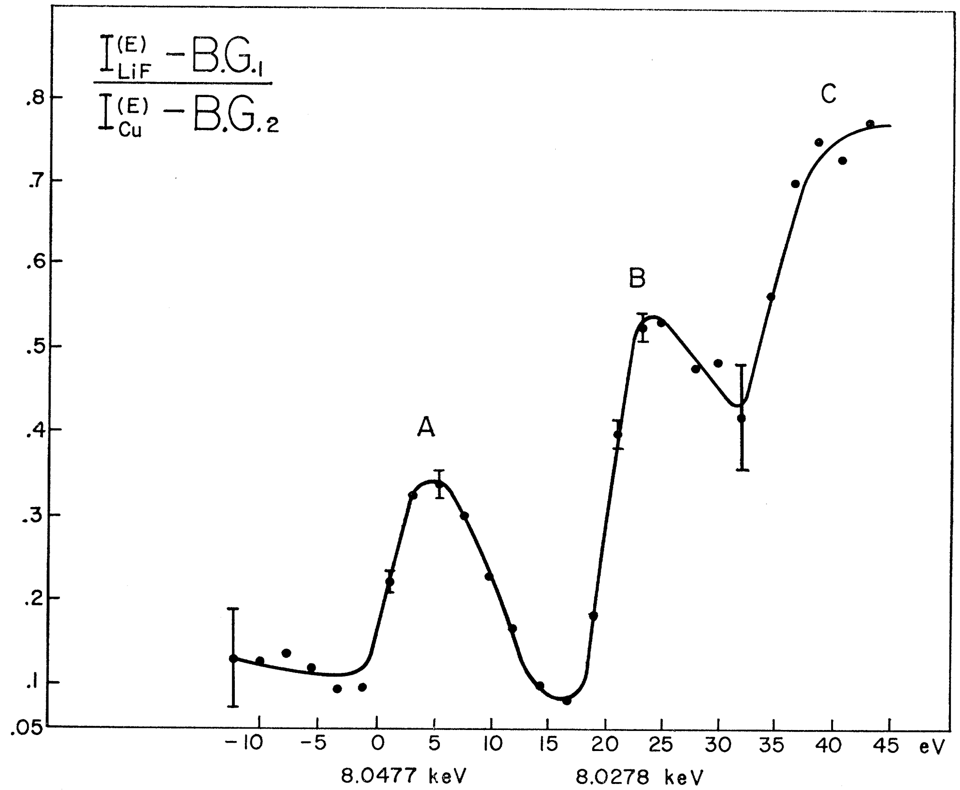

FIG. 3. Normalized incoherently scattered components.

in order to make a simultaneous evaluation of the data. This investigation will take a long time to carry out because of the very low counting rate and other experimental difficulties. The results should be very interesting, however, because they would yield some understanding of the effect, information about the effective mass, and other properties of *F* centers.

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