Compton profile measurements of lithium fluoride with 662 keV radiation

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Abstract

The isotropic Compton profile of lithium fluoride has been deduced from Compton scattering measurements on poly crystalline sample at gamma ray source energy of 662 keV. Here we report the experimental and theoretical Compton profile of lithium fluoride. We calculated directional Compton profile and their anisotropic effect using self-consistent Hatree-Fock wave functions employed on the linear combination of atomic orbital (HF-LCAO) approximation. The spherical average Compton profile has also been calculated and compared with the experimental results. The experimental results are found to be in good agreement with the HF-LCAO results and in qualitative agreement with Hartree-Fock free atom values.

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1. Introduction

The Compton scattering is a powerful tool for determining the momentum distribution of electron in condensed matter [1, 2]. The Compton scattered radiation is Doppler broadened because of the motion of target electrons. An analysis of this broadened line shape gives valuable information about the momentum distribution, n(p), of electrons through the Compton profile, $J(p_z)$, which is defined as

$$J(p_z) = \int\int n(p)\, dp_x\, dp_y \qquad (2)$$

Thus $J(p_z)$ is the one dimensional electron momentum distribution. The Compton profile is directly obtained from the spectral distribution of photons emitted by a monochromatic radiation source and scattered at a fixed angle. Increasing interest has been directed towards the experimental and theoretical determinations of isotropic Compton profiles and anisotropies in the directional Compton profile. The Compton profile studies in ionic solids, particularly alkali halides with NaCl structure, have been the subject of great interest both experimental and theoretically. Amongst all the alkali halides the fluorides have been theoretically predicted [3] to give larger anisotropy and it has been shown that the relative anisotropy of fluorides decreases as one goes from lithium to higher alkali atoms [4]. Lithium fluoride is the simplest of all typical ionic solid; it has a NaCl structure and ten electrons per unit cell. For that reasons it has been the subject of intensive theoretical investigation [5 - 8] and measurement [9 –11]. But lithium fluoride was the first alkali halide for which directional Compton profiles have been measured [9, 12]. The calculation of Beggren et al [11] using Kunz's crystal orbitals and series expansion orthogonalization agreed reasonably well with their experimental results. There are also other reasons for studying the lithium fluoride. The ground state may simply be viewed as a superposition of closed shell ions which overlap each other only very weakly. For theory lithium fluoride is therefore one of the simplest systems available since an extreme tight-self consistent model is likely to yield a highly accurate electronic structure. Thus lithium fluoride offer a beautiful opportunity for studying the various assumption which are incorporated in the Compton scattering technique, such as the impulse approximation, relativistic, and multiple scattering corrections. Still another reason for the present study is due to widespread use of LiF in thermoluminesecence dosimetry its properties are well studied and it is possible to compare the high-energy irradiation effects of other types of luminescence exitations. In contrast to the previous alkali halides, lithium fluoride is a more complicated material for the luminescence spectroscopy since its spectra, besides intrinsic self trapped exciton emission bands, consists of, usually, the bands assigned to radiation defects and trace impurities. The latter bands are typical for LiF because of its relatively high melting temperature. The understanding of the ground state electronic structure is necessary for the explanation of chemical bonding and hence various properties.

These interesting properties has tempted us to study in detail the electronic structure of lithium fluoride in comparison with other ionic solids using 661.6 keV gamma ray source. We have calculated the theoretical Compton profiles using Hartree-Fock wave functions employed on the linear combination atomic orbital approximations (LCAO) by CRYSTAL98 program and also to the best of our knowledge this is the first experimental study of the lithium fluoride using 662 keV gamma radiation. The experimental and theoretical results will be discussed in the following sections.

2. Experiment

In our experimental set up the scattering chamber is a single evacuative cylindrical chamber made of 5mm thick steel of length 2700 mm and diameter 510 mm. It has been made long to minimize the wall background signal. It is surrounded by lead walls of thickness 50 mm. The source is housed inside this chamber; the larger dimension of the chamber together with the scattering angle, which is fixed at predetermined value of about 168,° allows a good separation of the incident and scattered beams. In the present experimental arrangement the source used is ^{137}Cs of strength 2 Ci and a half-life of 30 years. The source is encapsulated in a steel cylinder of diameter of 16 mm and height 25 mm. The diameter and height of the active material are 11.5 and 18.5 mm respectively. The energy of the ^{137}Cs main line is 662 keV and it lead to less self-scattering per unit volume in the source material. The source is placed in a lead block and emitted gamma ray is collimated through a collimator of diameter 1.5 cm before being incident on the sample. The scattered beam is defined by collimators and leaves the chamber through an aluminium window 0.2 mm thick. To avoid the scattering from the air particles the scattering chamber is evacuated and the pressure is held below 0.1mbar. The design of the block and scattering chamber should include a precise determination of the scattering angle and the geometric contribution to the total resolution function. These become important in determining the accuracy of the data. High purity (99.99%) lithium fluoride powder was pressed to form a pellet of diameter 2.5 cm and thickness 0.11 cm and packing density 2.64 gm/cm^3. This pellet was mounted on a sample holder which is held vertically in Y- θ target table. The Y – theta target table is a Computer / pendant controlled system is placed inside the scattering chamber for positioning a wide range of materials with an extremely high degree of accuracy. The Y-theta target table simplifies crystal alignment, such a system has been installed, which additionally allows for a rotation of the sample about the goniometer axis without opening of the scattering chamber. The Y- theta target table has been designed such that by placing the material to be studied on the system, any position on Y and theta axis can be achieved by operating though a control system, Pendant and Man machine interface. So, the sample can be mounted to scatter the incident beam at fixed an angle with pinpoint accuracy by using this computer controlled Y-theta target table. The back-scattered gamma radiation was further collimated and detected by a thin high purity planar germanium detector of area 200mm^2. The momentum resolution of the detector, which depends on the detector properties and beam divergence, was about 0.41 a.u. (1 a.u. = 1.99289 × 10^{-24} kg m/s). Over 70,000 counts per channel were collected under the Compton peak having each channel calibrated as 0.144 a.u. To correct the background contribution, a separate measurement without any sample was taken, by keeping the source in position and subtracted the data from the measured spectrum point by point after being properly scaled to the sample real measurement time. The background radiation also consists cosmic radiation and bremsstrahlung radiation from photo electrons and Compton electrons ejected from the atomic shells of the sample material by the interaction with the incident photons. In this experimental arrangement the effect due to cosmic radiation was negligible in the detector and can be neglected. The Compton electrons can have a maximum kinetic energy of 476 keV for 662 keV incident photons which had not contributed in the energy range of our interest. The experimental data was then corrected for the sample absorption and energy dependence of the Compton scattering cross-section. The double scattering contribution was calculated at this stage by the Mento-Carlo simulation technique following the method of Felsteiner et al [13]. We have not considered here any correction for higher order scattering, but as discussed by Halonen et al [14], if the ratio of double to single scattering is a, then the fractional contribution due to higher than second order scattering turns out to be $\alpha = a^2/(1 - a)$. In this experiment the ratio of double to single scattering was found from Monte-Carlo simulation to be 3.0%, so the fractional contribution due to the higher order scattering was calculated as 0.093%. So, the contribution of higher order scattering is less than the statistical accuracy and has thus been neglected. The raw experimental data is shown in Fig. 1.

After considering all the correction factors mentioned above the results were then deconvoluted following the generalized least square method of Paattero et al [15], and then it was normalized to the total number of electrons obtained from the free atom Hartree-Fock values taken from Bigg's table which is equal to 5.865 electrons for lithium fluoride in the momentum range 0 to 7 a.u. Finally the experimental and HF-LCAO Compton profile values of LiF are shown along with Hartree-Fock free atom values in Fig. 2. The theoretical profiles have been convoluted with residual instrumental function.

3. Computational method

Theoretical Compton profiles for LiF are calculated using the CRYSTAL98 suite of program [16]. Crystalline orbitals were generated from self-consistent Hartree-Fock method applied to linear combination of atomic orbitals (LCAO). The calculation was performed at the characteristic crystal structure (rock salt) using the experimental lattice parameters (a = 4.0173 Å) according to Wyckoff [17]. Default tolerances for the calculations were used in the calculation, with reciprocal space integration being performed at 29 points in the irreducible wedge of the first brillioun zone. Calculation was performed with high quality all-electron basis sets, namely 6-1G [18] for Li^+, and 7-311G for F^- [19, 20]. All these basis sets have previously been optimized by the authors of CRYSTAL98 for the systems studied in this work.

The experimental isotropic Compton profile of lithium fluoride, HF-LCAO theoretical profile and Hartree-Fock free atom results obtained from Bigg's et al [21] after convoluting with the residual instrumental function of the experimental arrangement are shown in Fig. 2. The experimental and theoretical data agree well with calculated and free atom values.

The determination of Compton profile is one of the methods, which provides critical test to the band wave functions, it also provide important information on some features of the Fermi surface. Furthermore, the study of the anisotropy of the Compton profile can lead to the understanding of the nature of bonding in solids. The calculated isotropic Compton profiles of lithium fluoride for some selected values of q ranging from 0 to 7 a.u. are compared with experimental values.

Using the same Hartree-Fock wave functions employed in the LCAO calculation of isotropic Compton profile the directional Compton profile of lithium fluoride along the three principal directions, namely [100], [110] and [111] for q ranging from 0 to 7 a.u. were also calculated. This theoretical result of LiF were convoluted using the same experimental resolution function and normalization factor as used in the calculation of isotropic Compton profile. Differences of the Compton profile with respect to directions of the momentum transfer for LiF are shown in Fig. 3.

In Table 1 the experimental isotropic Compton profile of lithium fluoride, HF-LCAO theoretical profile and Hartree-Fock free atom results obtained form Biggs et al [21], after convoluting with the residual instrumental function of the experimental arrangement, are given. The experimental and HF-LCAO theoretical data agree well and free atom values agree well in the high-momentum region, as is also evident from Fig. 2. The isotropic Compton profiles for lithium fluoride for three principal directions are given in Table 2.

The theoretical Compton profiles involving the main symmetry directions are shown in Fig. 3 for lithium fluoride. From the same figure the effects of the shape of the Fermi surface also can be interpreted. The most pronounced effects can be found in the low momenta region. The structures originating from the directional dependence of the Fermi surface may be seen as well. It is seen that the high momentum anisotropy is mainly due to [100]. The fact that the [110]-[111] anisotropy is very small in the high momentum region is because both the directions cut the [100] bond at roughly the same angle, and thus to the first-order the bond look the same. Further for the difference Compton profile [111]-[100], in Fig. 3 a significant effect is seen for momentum values $q \leq 1.8$ a.u., where a pronounced minimum is observed around $q = 1.4$ a.u. However, the J[110] – J[111] curve has an amplitude roughly one half of the amplitude of the other two anisotropy curves. Thus, one can conclude that the crystal direction [100] could be a special one. This is the nearest neighbour (Li – F) direction, which has been found to be special one.

The differences (ΔJ(q)) between convoluted theoretical and experimental Compton profiles of lithium fluoride is shown in Fig. 4. It is seen that at lower momentum the deviation is more for free atom values compare to HF-LCAO calculations. At higher momentum region the agreement between experiment and free atom theoretical Compton profile are found to be good. The over all agreement between experiment and theory is seen to be much better for the HF-LCAO calculation, it is also evident from the Figs. 2 & 4. Excellent agreements, between experiment and HF-LCAO results and in the higher momentum region for free atom results, are found which provides confidence in this measurement and data analysis.

4. Conclusions

Experimental isotropic Compton profiles of lithium fluoride after all corrections together with the theoretical profiles after convoluted with the residual instrumental function are presented. The theoretical Compton profiles have been calculated by employing Hartree-Fock wave function on LCAO approximations. As can be seen from the Table 1 and Fig. 2 the experimental Compton profiles and HF-LCAO results are in close agreement and however the free atom profiles are only in qualitative agreement with experimental profiles of lithium fluoride. As a further check, the autocorrelation function B(R) were determine by transforming the Compton profiles with a fast Fourier transform (FFT) program. The results are shown in Fig. 5. The differences ($\Delta J(q)$) between convoluted theoretical and experimental Compton profiles of lithium fluoride is also showed that the over all agreement between experiment and theory is seen to be much better for the HF-LCAO calculation. At higher momentum region the agreement between experiment and free atom theoretical Compton profile are found to be good.

The directional Compton profiles of lithium fluoride were also calculated using the Hatree-Fock wave function employed on LCAO approximation. Anisotropy Compton profiles are shown in Fig. 3 and isotropic profiles along three principle directions are given in Table 2. It is observed that the shape of the Fermi surface represented by its different cross-sections leaves a clear mark on the difference Compton profiles. The particular shape of the Fermi surface has heavily influenced the anisotropies at low momenta.

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Figure Captions:

Fig. 1. Raw scattered spectrum of 1.1 mm LiF

Fig. 2. Total Compton profile of lithium fluoride. The full curve with solid symbol represents the experimental Compton profile. The full curve with open symbol represents Compton profile values based on the HF-LCAO method and the broken curve represents the Hartree-Fock Compton profile values.

Fig. 3. Theoretical anisotropy curves of the Compton profile of lithium fluoride. Line with circle represents the J[110] – J[100] anisotropy, the line with solid circle represents the J[111] – J[100] anisotropy and the line with square represents the J[110] – J[111] anisotropy respectively.

Fig. 4. Difference Compton profile of theory and experiment of lithium fluoride. Solid line represents the difference of HF-LCAO and experiment. Broken line represents the difference of Hartree-Fock free atom values and experiment.

Fig. 5. Autocorrelation function of lithium fluoride. The broken curve represents the experiment and solid line represents the HF-LCAO theory.