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HIGH ENERGY K X-RAY HYPERSATELLITES

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ABSTRACT

This study focus on the weak lines that appear on high energy side of the diagram lines which is called High Energy K X-ray hypersatellites i.e., a particular category of characteristic X-ray due to splitting of fine structure levels. It's resulted from the interaction of spin of an electron with the spin of the nucleus. They are emitted when an atom that has undergone a spontaneous transitions from the higher hyperfine level to the lower one to a radiation of $\approx 1.42 \times 10^9 \mathrm{Hz}$ frequency and $\approx 21\ \mathrm{cm}$ wavelength. Simultaneous double ionization in the K-shell and multiple ionization in the L-shell gives rise to K_α X-ray hypersatellites. These can be studied only by high energy resolution instruments like WDXRF spectrometry. Fundamental experimental procedures were outlined in this field by several workers due to different excitation modes. The theoretical models to predict their energies and intensities were developed. And also this review can show a clear discrepancy between theoretical and experimental results in the case of hypersatellites formation from different shells. In case of experimental instrumentation, WDXRF is the most accurate for determining the energy and intensity of X-ray hypersatellites. All sources of data was literature done by different scholars.

Keywords: X-ray hypersatellites; Energy Ratio; Intensity Ratio

1. INTRODUCTION

X-ray satellites emitted in the radioactive decay of double-vacancy states when the two initial vacancies are located in the same shell say K shell are of particular interest. Such satellites were named by Briand et al[1] as the hypersatellites. The denotation of the spectra as $K^h_{\alpha 1,2}$ and $K^h_{\beta 1,3}$ originate in the single electron K-spectator-hole transitions $1s^{-2} \rightarrow 1s^{-1}\ 2p^{-1}$ and $1s^{-2} \rightarrow 1s^{-1}3p^{-1}$, respectively. They were first investigated by ion excitation and later by photon excitation using X-ray tubes. High-resolution measurements of heavy-ion-induced K hypersatellites were found to represent a sensitive tool for studying the relativistic and QED effects in atoms[2].

More recently, it has become possible to investigate the double-K-shell photo ionization process by measuring the K hypersatellite X-ray emission of light and mid-heavy elements irradiated with intense synchrotron radiation beams[3]. In double photo ionization despite of the electron shake-off process, the electron knock-out process appears, in which the first ionized electron can kick out a second bound electron leading to formation of a double-K-vacancy state. As a consequence, double K-shell photo ionization can only result from electron-electron correlation effects[4, 5].

An atom with one of its shells empty is called a hollow atom. These satellites can give information on the energy level structure of the electron shell of the hollow atom. They can be produced using target bombardment by high energy heavy ions. But when electrons are ejected, the resulting hollow atom is very difficult to control so that using photo excitation by monochromatic synchrotron radiation is preferable. But photo excitation method produces small probability for the creation of a double K-vacancy and is a disadvantage.

Well resolved photo excitation studies of the hypersatellite lines are very rare because the creation probability of hollow atom via photo excitation is very small. For example, for a $Z \approx 30$ atoms, the cross-section for the production of hypersatellite lines is $\approx 10^{-4}$ of that of the $K_{\alpha 1,2}$ and $K_{\beta 1,3}$ diagram fluorescence lines. However, study of hypersatellites using photo ionization is also reported by Raju et al[6] for elements in the Z range 19-25. Studies of the formation mechanism and the electronic structure of hollow atoms give important new insight in to fundamental issues like inter and intra shell electron correlations, the effects of relativity, and the Breit interaction. In addition to their importance for basic atomic physics, hollow atoms have important applications for studies of e.g. the electronic structure of surface, or systems far from equilibrium[7]. Hollow atoms have even been proposed as a way of achieving population inversion and lasing in the hard X-ray range of wavelengths[8, 9].

Relativistic effects also play a major role in the above mentioned transitions, due to the involvement of the highly relativistic K shell. While the effect of Breit interaction on almost all atomic transitions is less than 1%, for the hypersatellite transitions this effect can be as large as 20% in heavy atoms[10]. Therefore the hypersatellite lines offer a very rare opportunity to study in detail the Breit interaction, one of the least studied of all atomic interactions. The experiment done for a formation of hollow atom by bombarding high energy heavy ion, the number of electrons ejected and the state of the resulting hollow atoms are very difficult to control[11,12,13]. In this experiment photon excitation by monochromatic synchrotron radiation, this allows the measurement of a clean and high resolution hypersatellite spectrum and gives the possibility to select the exciting photon energy accurately. The disadvantage of the photo ionization method is the small probability for the creation of a double K vacancy, rendering high intensity synchrotron radiation sources vital for these experiments. A hollow atom can be producing either a single[14] or a multiphoton[15] process. Two photons are used to excite two K electrons nearly simultaneously. In a single photon excitation experiment, which was the method used in this work, both electrons are excited as a result of a single photon absorption.

The hypersatellites in these studies were very poor both in intensity and resolution. Richard et al [16] were the first to study hypersatellites of Calcium by heavy ion bombardment using the crystal spectrometer. This was followed by the work of Yohkoawaya[17] who measured K^2L^n/KL^n intensity ratio of Nickel, Iron, Chromium and Titanium by ion bombardment. Briand et al [18] observed hypersatellites in Copper, Nickel and Iron in a study by electron bombardment. Keski-Rahkonen et al[19] studied hypersatellite spectra of Magnesium, Vanadium, Chromium, Manganese and Iron by photon excitation. Pure Copper K_α hypersatellite spectrum generated by photo excitation using Synchrotron radiation is reported by Diamant et al [3].

In general, experimental procedures, theoretical computations based on some models, and some investigations carried out in this field at various laboratories in the world are examined with special references to chemical effects and Z systematics.

2. EXPERIMENTAL PROCEDURES

The procedure, for utilizing WDXRF, adopted in recording the spectra is to introduce the sample in the sample holder and to record the intensities in 2θ steps of suitable angular intervals ranging from 0.01 to 0.05^0. In general, good statistics are maintained in counting. In each case, the experiment is repeated at least four times, using fresh sample each time.

The collected data are first smoothed using sliding least square fitting and then the spectra are corrected for background using an appropriate computer programmer. The deconvolution of the peaks and estimation of areas under different peaks is carried out by one of the codes like 'PEAK-FIT'[20] software version 4.11 of Systat Software Inc. Energy calibration of the crystal is done by taking spectra of some element standards and assuming the established values of the diagram lines and Calibration parameters are determined. Using these energies of the X-ray is determined.

The estimated intensities, however, do not represent the absolute intensities, as they have to be corrected for various effects. Since the present interest is on the relative intensities and the energy regions covered for each element for the satellites are very small, the final total correction on relative basis is small. Hence normally no corrections need be applied[21]. However, corrections like Self-Absorption in the Sample, Crystal Reflectivity, Window Absorption, and Efficiency of the Detector are applied, even though small, in the present investigations.

3. THEORETICAL COMPUTATIONS BASED ON SOME MODELS

Different theoretical models are developed for the computation of energy shifts of hypersatellites relative to their respective diagram lines. The theoretical explanation of hypersatellite energy shifts can be defined by using calculations like;

D-H-S calculations: - Chen et al[2] estimated the energy shifts of the Kα hypersatellites $K_{\alpha 1}^{h}$ and $K_{\alpha 2}^{h}$ from the K_α diagram line. They have completed the energy shift of K_α hypersatellites relative to the diagram line in the intermediate coupling scheme using Dirac–Hartee-Slater wave functions and incorporating the full Breit interaction and the final state splitting produced by the Coulomb and Breit interactions in addition to the electrostatic interaction. The Breit interaction operator used was

$$H_{Br} = \frac{-1}{r_{12}}[\overline{\alpha_1}.\overline{\alpha_2}\cos\omega r_{12} + (1 - \cos\omega r_{12})]$$

$$\overline{\alpha_1}.\overline{\alpha_2} = \text{Diracmatrices}$$

Where r_{12} = distance between the two interacting electrons

For atoms with double inner shell vacancies, the multiplet splitting can be found by evaluating the corresponding coupled two hole matrix elements of the electrostatic and Briet interaction operators. As closed shells do not contribute to the multiplet splitting[22] the splitting of the double hole states is determined by the coupled – two-hole states alone. The sum of the electrostatic and Breit operators is,

$$H_{CBr} = \frac{1 - \overline{\alpha_1}.\overline{\alpha_2}}{r_{12}}\cos\omega r_{12}$$

Hence, the energy matrix of the electrostatic and Breit operators between the anti-symmetrized j-j coupled two hole states, which can be separated in to direct and exchange matrix elements is,

$$\langle j_1 j_2 JM | r_{12}^{-1}(1 - \overline{\alpha_1}.\overline{\alpha_2})\cos\omega r_{12} | j_1 j_2 JM\rangle = D - E$$

Chen[2] et al have calculated these shifts with some gaps in the Z range of present interest. Values are available for Calcium (Z=20) and Manganese (Z=25). The values for the other elements are interpolated from the values given in the **Table 1**. The theoretical hypersatellite energy shift varies smoothly with Z in this region.

Table 1 Theoretical Energy shifts (in eV) of the $K_\alpha^h L^0$ hyper satellites with respect to the $K_\alpha L^0$ diagram line[23]

Atomic number	$K_{\alpha 1}^{h}$ — $K_{\alpha 1}$	$K_{\alpha 2}^{h}$ — $K_{\alpha 2}$
18	184.4	175.5
20	206.3	197.4
25	260.7	254.9
30	317.3	314.0
36	388.0	387.6
40	438.9	439.1
45	506.6	506.4

M.C.D.F.Caliculations: - Multi Configuration Dirac Fock calculations made in the intermediate coupling scheme takes in to consideration the relativistic effects. But Breit interaction is omitted from the final splitting calculations. The Breit operator is expressed as,

$$\overline{H_{Br}} = -\frac{1}{2_{r_{12}}}[\overline{\alpha_1}.\overline{\alpha_2} + \frac{1}{r_{12}^2}(\overline{\alpha_1}.\overline{r_{12}})(\overline{\alpha_2}.\overline{r_{12}})]$$

Where α_i Dirac matrices and r_{12} is the distance between the two interacting electrons. In the MCDF calculations only the average contribution of this Breit operator which is valid only in the long wave length limit[24] is taken into account. According to Chen[2] these calculations tend to overestimate the hyper satellite energy shifts.

DEPENDENCE ON MODE OF EXCITATION, CHEMICAL EFFECTS AND Z SYSTEMATICS

A review of the literature shows that experimental investigations were carried out on hypersatellites on low and medium Z elements (mostly up to Z=32). In these studies attention was paid to the aspects of dependence on mode of excitation[25, 26], chemical effect[27] and Z systematics. Related to this for any element and chemical compounds regarding to their atomic number and projectiles has different energy shift and intensity ratio. When we see the oxidation number of any pure element is zero while the oxidation number of compounds are higher relative to one the other and also even if the oxidation number increase or decrease there is no variation in energy shift while the value of the intensity ratio increase or decrease.

Dependence on Mode of Excitation: 1) Projectile Dependence of Energy Shifts: - The K_α satellite energies are listed in **Tables 2**. In the case of the Al satellites, no systematic variation of the peak energies with projectile atomic number could be discerned.

Table 2. K X-ray energies for Al, Cl and K[28]

X-ray peak	Initial state vacancy configuration	Al	Cl	K
		X-ray energy		
1	$1s^{-1}$	1486.6	2621.9	3312.9
2	$1s^{-1}2s^{-1}$ $1s^{-1}2p^{-1}$	1496.9(±0.1)	2640.2(±1.4)	3336.0(±3.2)
3	$1s^{-1}2s^{-2}$ $1s^{-1}2s^{-1}2p^{-1}$ $1s^{-1}2p^{-2}$	1507.9(±0.2)	2658.8(±1.8)	3357.1(±4.0)
4	$1s^{-1}2s^{-2}2p^{-1}$ $1s^{-1}2s^{-1}2p^{-2}$ $1s^{-1}2p^{-3}$	1521.3(±0.3)	2678.5(±2.2)	3379.1(±4.7)
5	$1s^{-1}2s^{-2}2p^{-2}$ $1s^{-1}2s^{-1}2p^{-3}$ $1s^{-1}2p^{-4}$	1535.1(±0.3)	2699.1(±2.0)	3402.3(±5.7)
6	$1s^{-1}2s^{-2}2p^{-3}$ $1s\text{-}12s^{-1}2p^{-4}$ $1s^{-1}2p^{-5}$	1549.9(±0.5)	2720.3(±2.8)	3427.5(±5.3)
7	$1s^{-1}2s^{-2}2p^{-4}$ $1s^{-1}2s^{-1}2p^{-5}$	_	2743.5(±2.7)	3452(±5.6)
8	$1s^{-1}2s^{-2}2p^{-5}$	_	2769.8(±3.6)	3481.2(±5.6)

The listed energies are the averages obtained for all runs with all projectiles and the indicated errors are root mean square deviations. The absolute error in the satellite energies for Cl and K are essentially the same as those given for Al. **2) Projectile Dependence of Intensity Ratio: -** Watson et al[28] studied how $K_\alpha L^n$ relative intensity varies with different projectiles in Al, Cl and K.

Table 3. Relative K_α X-ray satellite intensity for Al, Cl, and K using 1.7MeV/amu[28]

Target	Projectile	No. of L shell vacancy							
		0	1	2	3	4	5	6	7
Al	H	0.858	0.142	_	_	_	_	_	_
	He	0.664	0.285	0.051	_	_	_	_	_
	C	0.068	0.215	0.366	0.239	0.090	0.023	_	_
	O	0.045	0.128	0.312	0.293	0.168	0.053	_	_
Cl	He	0.680	0.320	_	_	_	_	_	_
	C	0.053	0.229	0.363	0.256	0.094	0.005	_	_
	O	0.028	0.133	0.304	0.316	0.170	0.049	_	_
K	He	0.765	0.235	_	_	_	_	_	_
	C	0.074	0.307	0.353	0.208	0.058	_	_	_
	O	0.045	0.185	0.316	0.271	0.142	0.042	_	_

From the **Table 3** in all Al, Cl and K targets the intensity under He projectile decrease as a number of vacancy increase; while directly correlated with atomic number. As we see under the **Table 3** in the case of C and O

projectile the value of intensity is not consistent when Z increases. **3) Chemical Effects: -** A noticeable chemical effect was observed in the case fluorine K. Ram Narayana et al[29]. **4) Z Systematics (Z Dependence of Relative intensities): -** Raju et al[6] measured energies and intensities of K_α hyper satellites and K_β satellites of the elements in the Z range 19-25 by photon excitation. The relative intensity of the K_α hyper satellite with respect to that of the K_α diagram line is related to the ratio of double K-shell to single K-shell ionization. This intensity ratio was plotted as a function of Z. It decreases smoothly and exponentially with Z. The K_β satellite relative intensity with respect to that of K_β diagram line was plotted as a function of Z. This also was observed to vary exponentially and smoothly with Z. **5) Z Dependence of Energy Shift: -** Raju et al[6] supplemented their data with those reported by other authors on K_α hypersatellite energy shift relative to the K_α diagram line in the Z range 12-30 and studied the variation of this with respect to Z. They found the relationship to be linear. They obtained from the plot of this Δ(E) versus Z, the following empirical relationship

$$\Delta E(K_\alpha^h) = -3.0 + 10.048Z$$

CONCLUSIONS

WDXRF has an auxiliary collimator mounted in front of the detector helps in improving the resolution, analyzing crystal spectrometer is effected and data acquired by a personal computer making use of Philips super QMS windows based software, a suitable voltage and current to operate easily, sample changers for fast data collection etc. used to determining the energy shift and intensity of x-ray satellites. Due to this instrument the production of a satellites have been registered for a double-vacancy states existing in the K-shells is called X-ray hypersatellites.

When the atom is doubly ionized in K-shell and if one of these holes is filled by transitions from outer shell K X-ray hypersatellites (weak lines) are emitted. These lines appear on high energy side of the diagram lines. Study of K X-ray hypersatellites provides information on the; intra atomic relations, excitation dynamics, relaxation and other factors influencing X-ray emissions.

Energies and relative intensities of hypersatellites of high Z-elements were determined by various researchers using crystal spectrometer by applying photon, electron, and ion excitation modes. Investigations were carried out to examine chemical shifts, it was found that relative intensities are susceptible to the chemical environment but energies are not much affected by it.

Generally, from different literature, studies were not cover an energy from small Z-elements which have smaller wavelength of the satellites. So that the existing few studies about hypersatellites can be extended to cover all elements. Because a number of synchrotron facilities are being developed throughout the world; using these tunable hard X-ray sources, energy dependence of these processes can be studied more efficiently.

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