X-Ray Fluorescence as an Imploded Shell Diagnostic

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Abstract

When an imploded target shell is irradiated from within by hot-core x-ray emission, photoionization produces inner-orbital vacancies in atoms doped into the shell. Atomic electrons then fill these vacancies, resulting in characteristic K_α line fluorescence in the x-ray spectrum.[1] Due to the sensitivity of the amount of photoionization to the shell areal density, this fluorescence is a potential shell compression diagnostic. To explore this diagnostic concept, a simple model was developed to simulate the spectrum of a doped target shell.[2] The goal of this project was to compare this model with spectra from the detailed radiation-transport code Spect3D[3] for aluminum-doped target shells. The detailed simulation is a test of the assumptions of the simple model, such as optically thin K_α emission from the target shell. A discrepancy was found, suggesting most plausibly that the Spect3D atomic model is not consistent with the cold-metal fluorescence efficiency for aluminum used in the simple model. Further work is needed to determine whether the atomic physics model used in Spect3D is correct.

Introduction

At both the Laboratory for Laser Energetics and the National Ignition Facility (NIF), inertial confinement fusion (ICF) experiments are being carried out where targets are irradiated, causing them to implode.[4] NIF targets typically consist of an outer shell of germanium-doped plastic (CH) and an inner shell of cryogenic DT. Once peak compression has been reached, one obtains a compressed shell which is extremely high in density and a hot inner core which is low in density. With sufficiently high temperatures and densities achieved in NIF experiments, the target is expected to experience ignition, a condition where the energy production from fusion is substantial enough to be self-sustaining. One example of a diagnostic to measure the shell areal density, a measure of target shell compression implosion performance, is x-ray spectroscopic observation of the emission of x-ray fluorescence.

Shell fluorescence is the x-ray line emission that follows inner-shell photoionization of dopant atoms by x-rays emitted from the hot core.[1] This process is demonstrated in Figure 1. Typically, these appear as 2p-1s emission lines also known as K_α emission. The x-ray radiation emitted by the hot core of the target passes through the imploded target shell on its way to the spectrograph, creating an absorption spectrum at the detector. A relationship between the amount of measured K_α fluorescent emission and the amount of measured photoionization absorption seen in the target spectrum has been developed, based on a simple shell model.[2] If the model is upheld, this relationship can be used to infer the shell areal density. Complex radiation transfer codes can be used to validate this relationship between fluorescence and photoionization absorption and to ensure that the relationship between the amount of K_α fluorescence and the amount of photoionization absorption exhibited by the target shell is understood. Once this is

confirmed, this relationship becomes the basis of using photoelectric fluorescence as a diagnostic on future experiments on the NIF.

Overview of Fluorescence

In experiments currently occurring on the NIF,[4] the target consists of an outer shell of CH polymer, which is often doped with germanium. In order to investigate the atomic processes occurring inside the target shell using our detailed atomic model, it is necessary to use a simpler element as a surrogate for the germanium dopant, like aluminum because the Prism atomic model for germanium fluorescence is not yet complete.[1] Figure 1 shows schematically an imploded target similar to the ones used in NIF experiments, except that the shell is aluminum-doped hydrogen. When a target is irradiated, the target implodes, and the hot inner core emits x-rays which irradiate the outer shell of the target. This energy from the core interacts with the atoms of the dopant in the shell causing fluorescence to occur. The spectral distribution of the inner core emission is approximated by the expression

$$P_{core}(h\nu) = P_0 \frac{(h\nu)^3}{e^{h\nu/kT}-1}, \quad (1)$$

corresponding to a Planck or blackbody spectrum,[5] where $h\nu$ is the photon energy, kT is the core temperature, and P_0 is a constant. As this core emission passes through the target shell, some is absorbed and some is re-emitted as fluorescence.

The competition between various atomic processes, specifically spontaneous emission and autoionization, is a characteristic of the target dopant material and determines how much fluorescence occurs.[6] Photoionization absorption is the process by which some of the incoming x-ray photons from the hot core are absorbed when they photoionize K-shell electrons of the

atoms in the target shell (See Figure 2). Once the K-shell vacancy has been created, there are two possible atomic processes which can occur. First, the atom can be autoionized, which involves the internal emission of a photon that ionizes an outer shell electron. This can occur because the gap between the two energy levels in the transition is bigger than the energy required to ionize the electron. On the other hand, the vacancy can be filled by an outer shell electron along with the escape of a photon. If the vacancy is filled by an L-shell electron, by far the most likely of the possible radiative emission processes, then the emitted photon is a quantum of K_α fluorescence. The probability that the K-shell vacancy is filled by spontaneous K_α emission is expressed as the fluorescence efficiency, ω_{K_α}. Conversely, the probability that the vacancy is filled by autoionization is equal to $1 - \omega_{K_\alpha}$.

The fluorescence efficiency is the relative probability of spontaneous emission following the formation of a K-shell vacancy, which can be written in the form

$$\omega_{K_\alpha} = \frac{A_r P_{2p}}{A_r P_{2p} + A_a P_{2p}(P_{2p}-1)} . \quad (2)$$

In this expression, the probability that a 2p sublevel electron decays, $A_r P_{2p}$, is directly related to P_{2p}, the number of electrons in the 2p level. The competing probability that a 2p electron decays and transfers its energy to another 2p electron is equal to $A_a P_{2p}(P_{2p} - 1)$. The product $P_{2p}(P_{2p} - 1)$ represents the number of possible interacting electron pairs that can undergo the autoionization process after an inner shell vacancy is formed.[7] Figure 2 illustrates schematically the interaction between the atomic processes which are significant to fluorescence. Fluorescence appears as a measurable quantity of emission at the detector, and this emission is related to the quantity of photoionization absorption that occurs.

PrismSPECT and Spect3D

To study the conditions occurring during a target shell implosion, complex radiation transfer codes have been developed to model the irradiation of a compressed target shell by hot core radiation. One such program is Spect3D.[3] This code models the transfer of radiation and electrons between atomic sublevels by solving the implicit simultaneous rate equations which approximate the transition rates among the atoms of the target shell. These rate equations include all the atomic kinetics processes which are necessary to simulate fluorescence, including autoionization, photoionization, photoexcitation, and spontaneous emission. In addition, the codes include a large database of atomic levels and ionization species which permits the investigation to be as detailed as necessary. Another feature which is significant in demonstrating fluorescence is the ability to turn on and off certain atomic kinetics processes.

Figure 3 shows an x-ray spectrum produced by a Spect3D simulation. In this case of interest, a shell of 150 μm inner radius and 25 μm thick consists of a 1 g/cm^2 mixture of hydrogen with a 1%, by atom, concentration of aluminum. At the center is a small blackbody source of temperature 3 keV. It is made opaque enough to produce a blackbody spectrum, identical to Eq. (1), over the entire simulated spectral range, up to $h\nu = 10$ keV. Figure 3 shows an x-ray spectrum which exhibits both K_α fluorescence and photoionization absorption. The other feature which appears on the spectrum is a set of 1s to 3p sublevel absorption lines. The spectrum also shows distinctive lines for each species of the dopant in the target shell. This represents a normal simulation where all relevant processes are considered. Figure 4 is another Spect3D spectrum; however, this spectrum was generated with a zero rate coefficient for photoionization. As seen on the spectrum, no clear fluorescence lines appear. This demonstrates that Spect3D models fluorescence as a direct result of K-shell vacancies created by

photoionization of K-shell electrons. The implication of this is that Spect3D can be used to investigate fluorescence.

Simple Spectrum Model

Although the complex spectra simulated by Spect3D are helpful, it is useful to develop a simpler model in order to better understand and to interpret the data. Such a model has been developed.[2] The emitted continuum spectrum can be modeled by the equations

$$P(h\nu) = P_0 \frac{(h\nu)^3}{e^{h\nu/kT} - 1} e^{-\tau_L (\frac{h\nu_K}{h\nu})^3}; h\nu < h\nu_K$$

and

$$P(h\nu) = P_0 \frac{(h\nu)^3}{e^{h\nu/kT} - 1} e^{-(\tau_K + \tau_L)(\frac{h\nu_K}{h\nu})^3}; h\nu \geq h\nu_K \,, \qquad (3)$$

where P_0 is the intensity coefficient from Eq. (1), $h\nu$ is the photon energy, kT is the core temperature, τ_K is the K-shell contribution to the opacity at the K-edge, τ_L is the L-shell contribution to the opacity at the K-edge, and v_K is the photon energy at the K-edge, the energy threshold of K-shell photoionization. The Spect3D simulation output is provided as a set of spectral data points with the form ($h\nu, P$). We treat this synthetic spectral data as actual measured data. We fit the simple model to this data in order to estimate four unknown parameters, the core temperature, the initial intensity, the K-shell opacity and the L-shell opacity. Using four Spect3D data points, these four unknown parameters can be solved for, giving the user another set of information about the conditions occurring in the imploded target. Figure 5 demonstrates a Spect3D generated spectrum along with the four point model, showing in

particular the drop in the spectrum at the K-edge. The size of this drop is directly related to the shell areal density and to the amount of fluorescence. Therefore, a measurement of the fluorescence provides an independent measurement of the areal density.

Relating Fluorescence and Shell Areal Density

The shell areal density is a characteristic of an implosion which is vital to the success of the implosion. Fortunately, a relationship between spectrum and imploded shell parameters has been found[2] which allows the areal density to be inferred. This relationship can be expressed by the equation:

$$\frac{\int P(h\upsilon)_{K_\alpha} dh\nu}{\Delta P_{K-edge}\, h\nu_{K_\alpha}} = \omega_{K_\alpha} F(\tau, kT/h\nu_{K-edge}) \ . \qquad (4)$$

In Eq. (4), $P(h\upsilon)_{K_\alpha}$ is the spectral power of the K_α emission, ΔP_{K-edge} is the change in spectral power at the K-edge, ω_{K_α} is the fluorescence efficiency, and the function $F(\tau, t)$ is a slowly varying function of optical thickness τ and the core temperature t in units of the K-edge energy, $t = kT/h\upsilon_{K-edge}$. This equation says that the ratio of the number of K_α photons that are emitted to the number of photons that are absorbed creating K-shell vacancies is equal to the K_α fluorescence efficiency multiplied by a correction factor given by the function $F(\tau, t)$ of the dimensionless variables τ and t. This function is of order unity, and relates the number of photoionization events to the amount of energy absorbed by photoionization. It is primarily determined by the temperature of the core continuum spectrum and by the optical thickness of the shell. This function has been obtained for the case where the K_α photons escape freely and where the shell thickness is small, relative to its radius.

The function $F(\tau, t)$ is given by the expression

$$F(\tau,t) = \int_1^{\infty} \frac{(1-e^{-\tau/x^3})}{(1-e^{-\tau})} \frac{(e^{1/t}-1)}{(e^{x/t}-1)} x^2 dx \quad . \tag{5}$$

This was provided by Dr. Epstein[2] and has been plotted in Fig. 6 as a family of functions of τ for several values of t. The point corresponding to the case shown in Fig. 5 is indicated by the small circle. The detailed derivation of this expression was a "given" for the purposes of this project and is not repeated in this report. What it represents is the spectral integral of the photoionization probability for a photon passing through the shell, weighted by the photon spectral distribution obtained from the photon energy distribution given by Eq. (1). The integrand is normalized to unity at the K-edge, ($x = 1$ or $h\nu = E_{K-edge}$).

One relationship on which the above model is based is

$$\frac{(P_{K-edge} - \Delta P_{K-edge})}{P_{K-edge}} = e^{-\tau_K}, \tag{6}$$

where τ_K is the optical thickness at the K-edge due to K-shell photoionization, P_{K-edge} is the power at the top of the K-edge, and ΔP_{K-edge} is the drop in the flux at the K-edge.[5] The areal density $\rho\Delta R$ of the shell is related to τ_K according to

$$\tau_K = \mu\rho\Delta R \quad , \tag{7}$$

where μ is the contribution to the mass absorption coefficient due to the K-shell photoionization of the dopant in the target shell. Using the tabulated known value of μ for aluminum,[8] Eq. (7)

gives a result completely consistent with the simulated spectrum in Fig. 5, indicating that Spect3D is calculating the photoionization portion of the fluorescence process correctly.

An important goal of this project has been to validate Eqs. (4) and (5) using the simulated spectrum generated by Spect3D and shown in Fig. 5, with its K-edge continuum drop and K_α emission. We obtain $\int P(h\nu)_{K_\alpha} dh\nu = 1.37 \times 10^{16}$ erg/cm^2/sec from the spectrum by subtracting the continuum fit shown as the red line in Fig. 5 to isolate the K_α emission, which is then integrated numerically using the trapezoidal method in a simple computer program. The drop in the continuum at the K-edge $\Delta P_{K-edge} = 1.69 \times 10^{15}\ erg/cm^2/sec/eV$ was also measured directly from this simulated spectrum. The fluorescence efficiency of Al, $\omega_{K\alpha}$, is known to be 0.0387[6] and the photon energy of K_α, $h\nu_{K_\alpha}$, is 1487 eV. For these values, Eq. (4) is consistent with a value of F(τ,t) of 0.141, but one obtains from the model the value $F(\tau, t) = 0.720$ for kT = 3 keV and $\tau = 0.757$. The result, then, is that Spect3D predicts about 20% of the K_α emission expected by the model.

Even with the simplicity of the fluorescence model, its underlying assumptions are reasonable, so this factor-of-5 disagreement is larger than expected. The most plausible explanation for this discrepancy, and the most promising direction for future work, is that the cold-metal value for the fluorescence efficiency of aluminum, used by the model, is not being reproduced by Spect3D for the conditions of the experiment. There is no reason to doubt the completeness and accuracy of the Spect3D atomic model for Al, however. In retrospect, estimates by R. Epstein and B. Yaakobi[9] suggest that electron collisional and radiative recombination effects, which are included in the Spect3D atomic model, might reduce the effective fluorescence efficiency of lower-Z elements like aluminum, perhaps enough to account

for this discrepancy. The additional non-radiating processes would add to the denominator in Eq. (2). This disagreement is not encouraging, but it is a preliminary result that does not yet rule out using fluorescence as an areal density diagnostic for imploded targets and invites further work in that direction.

Conclusion

Fluorescence is the x-ray line emission which follows inner shell photoionization. This fluorescence emission is sensitive to the shell areal density, allowing a relationship to be formed between the amount of fluorescence and the amount of photoionization absorption. Spect3D provides a detailed model for the irradiation of an imploded target shell by emission from a hot inner core that includes all of the relevant atomic processes involved in fluorescence as a part of its atomic rate equations. Using this program, it is possible to demonstrate and study a simple model for the radiation from an imploded target shell. In particular, simple theoretical relationships which have been developed have been tested using simulated data provided by the Spect3D program. For one example, a factor-of-five discrepancy was found between Spect3D and the simple model. For now, the fluorescence of aluminum cannot be considered reliable without further simulation and without investigation into the underlying atomic physics.

The fluorescence from a germanium shell additive has been measured in several implosions on the NIF, however, and has been found to be consistent with the measured continuum spectra, according to the simple model, including Eq. 4 and Eq. 5, modified for the core emission spectrum at the much higher energies where germanium is photoionized. A proof-of-principle calculation for germanium fluorescence, similar to the one for aluminum carried out

in this project, would provide valuable insight into the ongoing implosion experiments on the NIF.

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References

1. J.J. MacFarlane, I.E. Golovkin, P.R. Woodruff, "Modeling of Inner-Shell Line Emission from Cu Targets Heated by Short Pulse Lasers," Prism Computational Sciences, Inc. Madison WI.
2. R. Epstein, private communication; R. Epstein *et al.*, Bul. Am. Phys. Soc. **56**, No. 12, 67 (2011) ; S. Regan et al., Phys. Plasmas.
3. Prism Computational Sciences, Inc., Madison, WI; J. J. MacFarlane *et al.*, HEDP **3**, 181 (2007).
4. Lois H. Gresh, Robert L. McCrory, and John M. Soures. *Inertial Confinement Fusion: An Introduction*. Laboratory for Laser Energetics: Rochester, NY. 2009; S.H. Glenzer et al., Plasma Phys. Control. Fusion 54, 045013 (2012).
5. S. Chandrasekhar, *Radiative Transfer*, Dover Publications, New York, 1960; R. Epstein, "The Equation of Transfer," personal manuscript - private communication, 2011.
6. J. H. Hubbell et al., J. Phys. Chem. Ref. Data, **32**, 339 (1994).
7. E. J. McGuire, Phys. Rev. **185**, 1 (1969).

8. J. H. Hubbell and S. M. Seltzer, on-line NIST Physical Measurement Laboratory report NISTIR 5632, www.nist.gov/pml/data/xraycoef/index.cfm.

9. R. Epstein and B. Yaakobi, Phys. Rev. **A 44**, 5111 (1991).

Figures

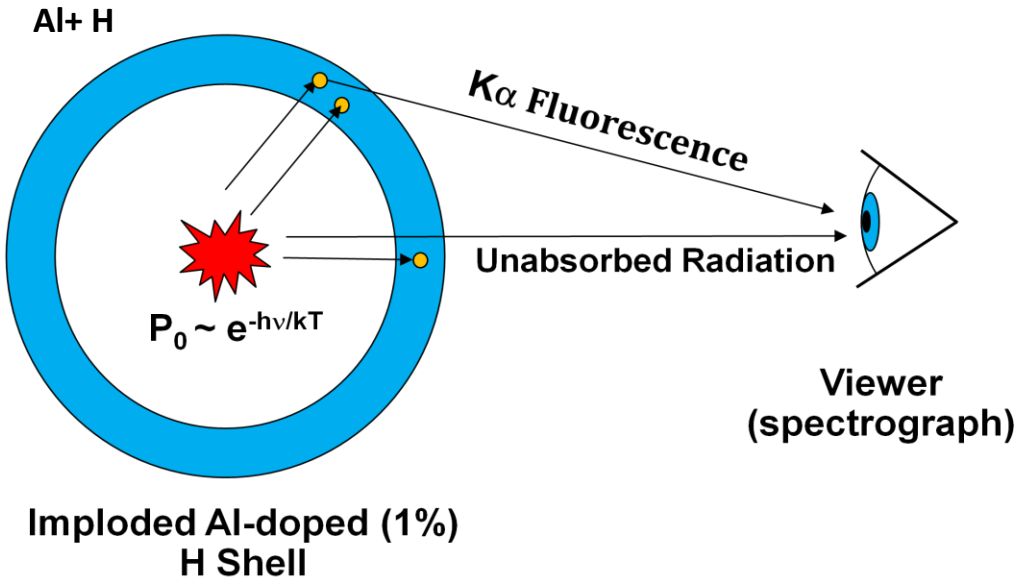

Fig. 1. An imploded target shell doped with aluminum. The target shell is being irradiated by x-rays from the hot core. These x-rays interact with the atoms in the target shell as they make their way to the detector.

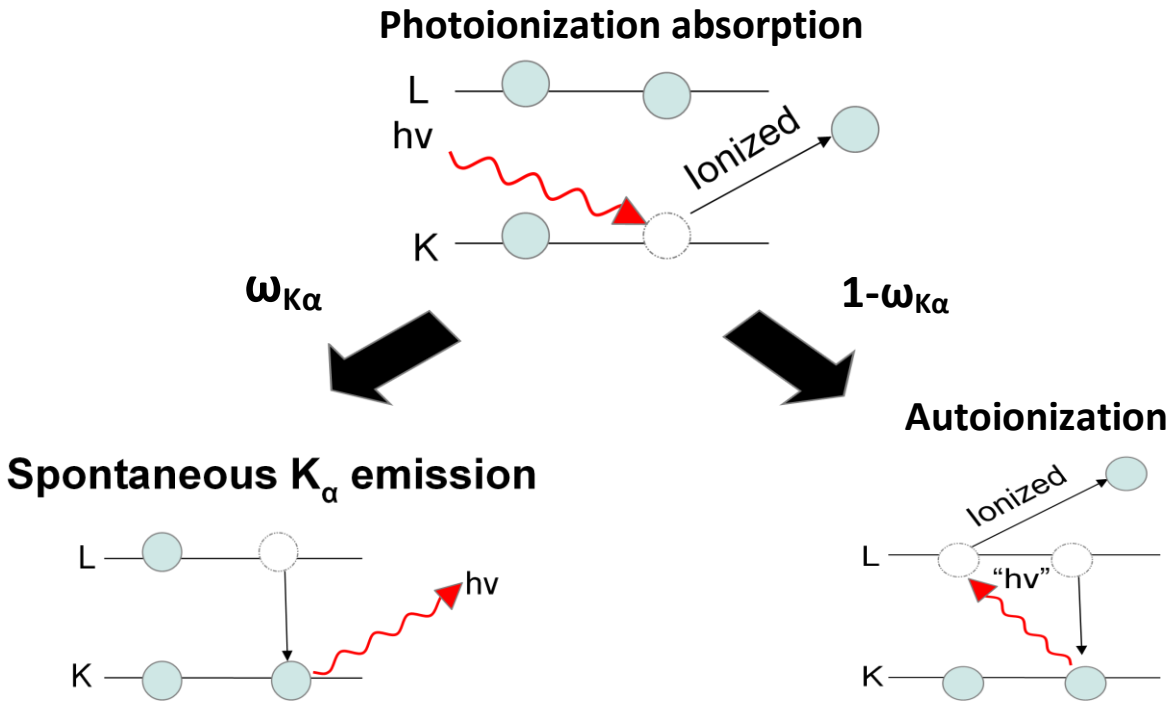

Fig. 2. Diagrams of the simple atomic processes involved in fluorescence. Photoionization, spontaneous emission, and autoionization are all shown. The probabilities, $\omega_{K\alpha}$ and $1-\omega_{K\alpha}$, of each process occurring after photoionization are also shown. The L shell has a higher electron energy level than the K shell.

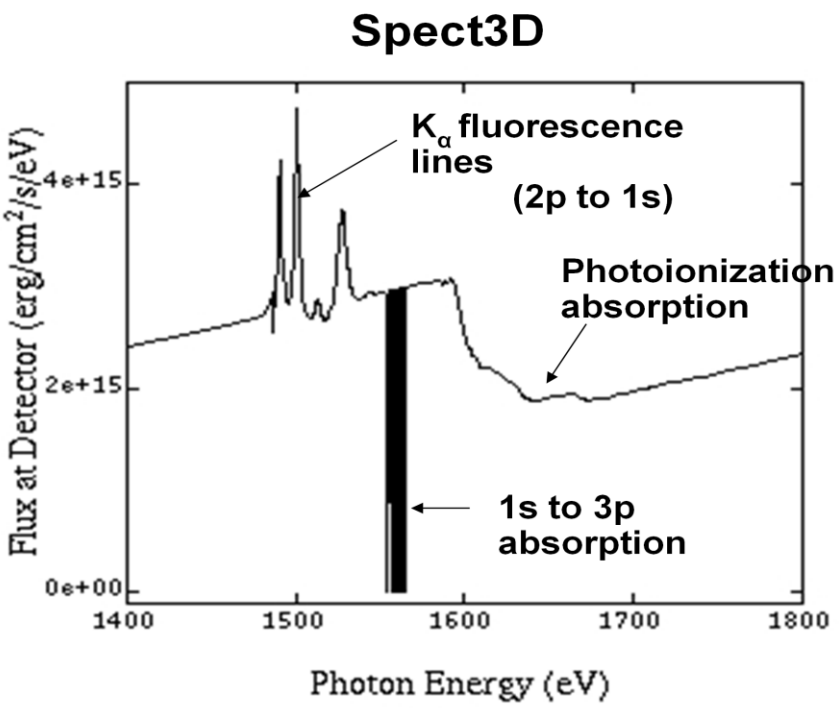

Fig. 3. A Spect3D simulated spectrum. The spectrum graphs photon energy vs. flux at the detector. The spectrum exhibits fluorescence lines, photoionization absorption, and 1s to 3p absorption lines.

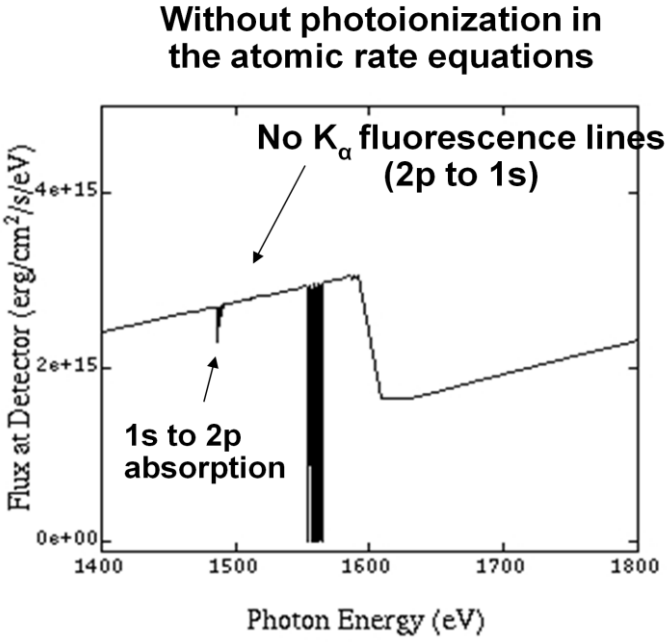

Fig. 4. A Spect3D simulated spectrum. This simulation was done without photoionization in the atomic rate equations which are solved by the complex radiation transfer code.

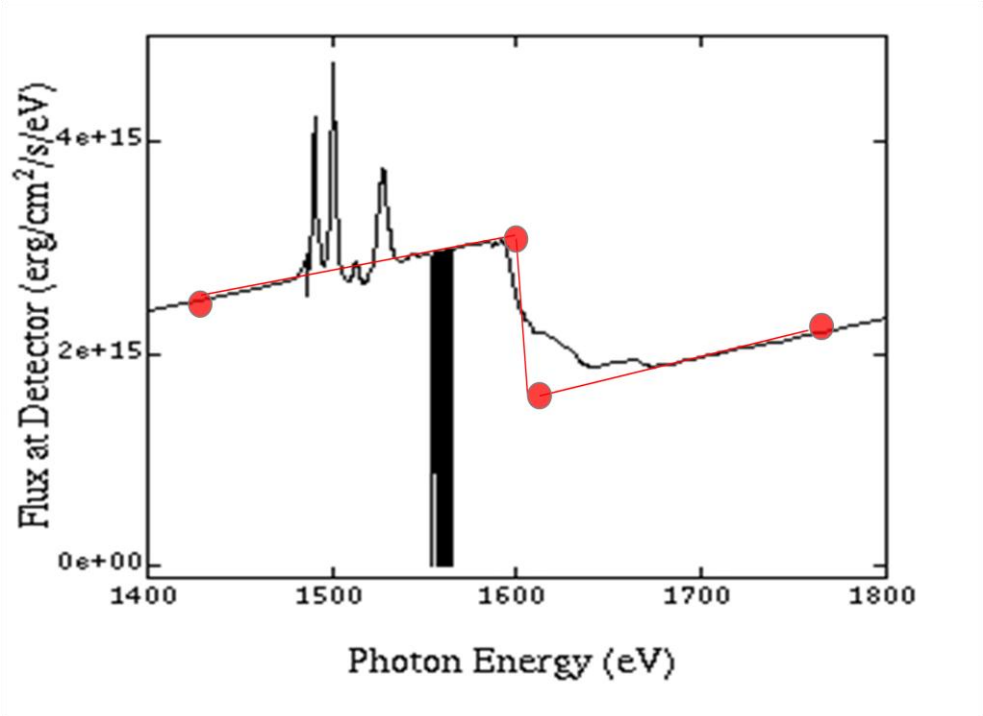

Fig. 5. A spectrum demonstrating the four-point model. This spectrum demonstrates the way in which a model of the emitted spectrum can be developed by extrapolating four data points from the Spect3D data.

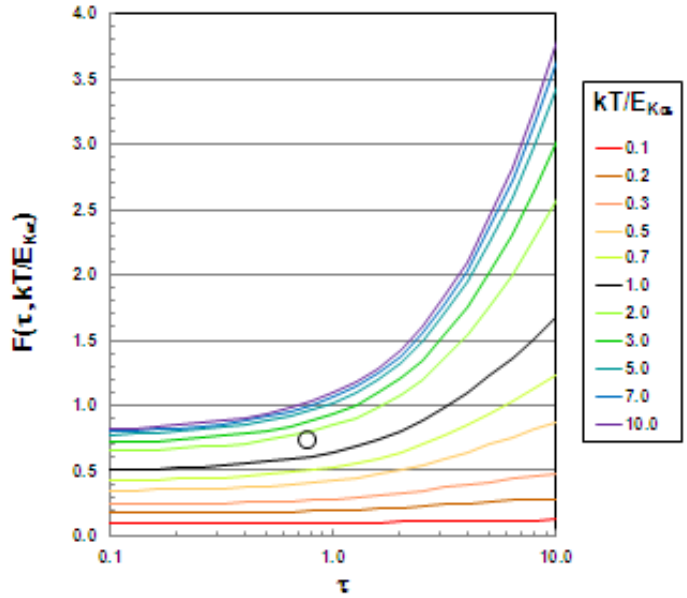

Fig. 6. The fluorescence response function plotted as a function of the shell optical thickness for various values of the source temperature in units of the K-edge energy. These results were obtained for the Planck continuum source created in the Spect3D simulation. Near the point $\tau =$ 0.757 and $kT/E_{K\alpha} = 1.92$, indicated by the small circle, the value of the function 0.720 varies slowly with these parameters.