Soft X-Ray Fluorescence from Particulate Media

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We introduce a Monte Carlo ray-tracing code for soft X-ray fluorescence in particulate media. We use the code to investigate the effects on absolute fluorescence line intensities and relative line ratios due to the medium porosity, incident spectrum, and particle size distribution as a function of observation geometry. In particular, we assess the differences between the results given by the simulations and by the analytical fundamental parameters equation.

INTRODUCTION

Soft X-ray fluorescence spectroscopy is a well-established method for elemental analysis of solid matter in laboratory conditions. The measured samples can be prepared for optimal usage, and the brightness and the spectrum of the X-ray source can be controlled to a great accuracy. However, when applied to remote observations of planetary surfaces by space probes, difficulties arise.

The first set of difficulties are due to the fact that the main source for the X-rays, the Sun, is highly variable and unpredictable. The shape and intensity of the X-ray spectrum varies in short timescales, and must be measured simultaneously with the actual fluorescence observations.

The second set of difficulties are inherent to the measured target, i.e., the planetary surface. The surfaces of terrestrial planets and most asteroids are rough, and mostly covered by regolith. The absolute fluorescence line intensities are sensitive to the surface properties such as the particle size distribution, particle packing density, and large-scale surface fluctuations. Furthermore, the effects on absolute line intensities are directly reflected on the ratios of the fluorescence lines, which are used in the elemental analysis of the medium.

Therefore, in order to do elemental analysis using remote X-ray fluorescence observations, it is necessary to know the expected magnitude of the volume and rough-surface effects for the used observation geometries. This knowledge will allow for a realistic assessment of the applicability of the simplified analytical models, such as the fundamental parameters equation (FPE), and will help to constrain the errors when using these analytical models to infer elemental abundances from the observation data. Secondly, a realistic model for the fluorescence output could be used in the future to deduce the regolith properties from the observational data, i.e., fluorescence observations in varying observation geometries can yield information of the surface structure.

Laboratory measurements of the effects on soft X-ray fluorescence due to varying particle size distribution and observation geometry has been carried out by Näränen et al. [1, 2]. An excellent introduction to the theory is given by Clark and Trombka [3], and in-depth reviews of the history of remote soft X-ray observations are given in [2, 1].

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SOFT X-RAY FLUORESCENCE SIMULATION

The simulation code is written in Fortran2003 and uses OpenMP for threading. The simulation data is stored using the netCDF-format, a machine-independent scientific data format, and readable using the standard netCDF tools. The xraylib-package [4] is used for the photoionization coefficient data, as well as for the other atomic data. The analysis of the simulation data is carried out using Python and SciPy.

The particulate media generation is carried out with a dropping-based packing method, described in [5]. Briefly, 5×10^6 spherical particles with radii following the given size distribution are packed into a given packing density inside a rectangular container of edge width w. The edge width is much greater than the mean particle size in order to make the scale of the inhomogeneities produced by the porosity small compared to the spatial extent of the packing.

We model macroscale surface roughness of several mean particle diameters and greater using two-dimensional random fields as described in [6]. The packing is intersected with a random field following Gaussian correlation (Gc) or fractional-Brownian-motion statistics (fBm), and the particles above the random field are removed. The random fields used are functions of two parameters: the standard deviation of heights σ and a model-specific parameter describing the horizontal roughness statistics. The fBm-fields are parametrized by the Hurst exponent H and Gc-fields by the correlation length ℓ.

Fluorescence ray tracing

The X-ray fluorescence ray tracing follows closely the standard ray-tracing principles. A small departure from the basic ray-tracing techniques is taken by calculating the first-order fluorescence signal from all fluorescence lines simultaneously.

For a ray R with radiance I, energy E, position vector $\mathbf{P}$, and direction vector $\hat{\mathbf{D}}$, we compute the mean free path γ from the total extinction coefficient μ_t. The simulation for a single ray begins by searching for the first medium-ray intersection point for the incident ray. Next, a random path length s is drawn from the exponential distribution $s \sim \exp^{-\gamma}$, and the ray is traced to a new position $\mathbf{P} = \mathbf{P} + s_{in}\hat{\mathbf{D}}$, where s_{in} is the distance traveled inside the particles.

Now, the first-order fluorescence signal for fluorescence line l emerging from the point $\mathbf{P}$ inside the medium to a given direction is

$$I_{\mathrm{f,l}} = \frac{I}{4\pi} F_\mathrm{l} f_\mathrm{f} f_\mathrm{e,l} \exp^{-s_\mathrm{out}\mu_\mathrm{t}}, \tag{1}$$

where F_l is the fluorescence yield of the line, f_f and $f_\mathrm{e,l}$ are ratios calculated from the photoionization coefficients, s_out is the distance traveled inside the particles from $\mathbf{P}$, and obtained by tracing a ray until it exits the media. The variables F_l, $f_\mathrm{e,l}$, and μ_t are vectors, and can be precomputed into a lookup table for efficiency.

RESULTS

Absolute line intensities

First we consider the absolute fluorescence line intensities from simple one-element media. We investigate simulated Fe and Ca Kα-line fluorescent signal as a function of varying par-

ticle size distribution models, mean particle sizes, medium packing densities, and incident spectra. We chose two particle size distribution models, monodisperse (constant radius) and uniform, parametrized by the mean particle size (r = 5, 10, 50, 100, and 250 μm). We used two spectrum models, a 9 keV line source and a continuous Ti-tube spectrum. The Ti-tube spectrum was chosen to match closely to the spectrum of the Ti-tube source used in the laboratory measurements by Näränen et. al [1, 2]. Finally, we selected three packing densities ($\rho = 0.20, 0.35, 0.50$).

In Fig. 1 we show the absolute Fe Kα-line intensities for the Ti-tube spectrum, uniform size distribution, three packing densities, and five mean particle radii, together with the results from FPE modelling. The main results are:

- Deviations from the FPE results can be large. Media with low ρ and large r can produce half of the FPE signal for certain observation geometries.
- The mean radius r is one of the most important factors affecting the strength of the absolute line intensities. The effect of changing r is most notable at the transition region where r is close to the mean free path γ.
- The role of the size distribution is small, especially outside the transition region ($r << \gamma$ or $r >> \gamma$). This is because the fluorescence takes place increasingly at the surfaces of single particles for $r >> \gamma$, and for $r << \gamma$ the radiation travels through many particles before ionization.
- Large r values yield a linear trend for $\theta_e > 15°$, and a nonlinear brightening is visible at small θ_e. The nonlinear brightening becomes more pronounced with decreasing ρ, and the curves are nearly linear for the whole θ_e-space for large r and large ρ.

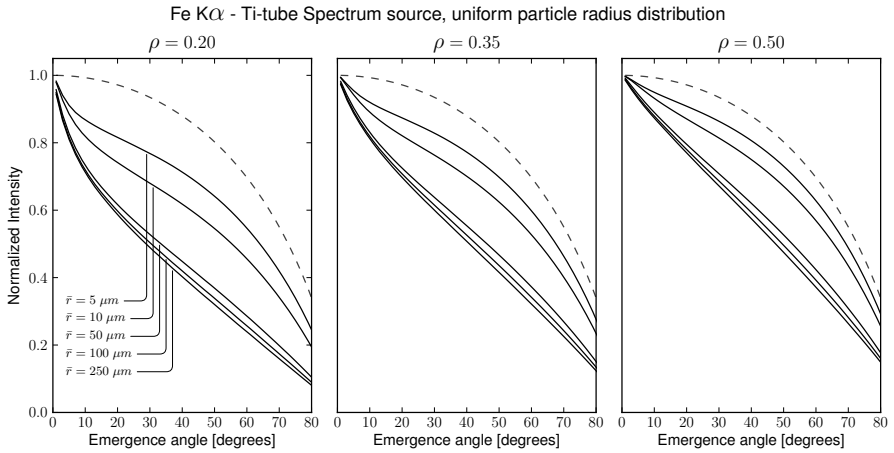

Figure 1. Fe Kα-line intensities as a function of θ_e for $\theta_i = 0°$, Ti-tube source, uniform size distribution, three ρ's, and r's. The results by FPE are shown as dotted line.

Fluorescence line ratios

Next, we focus on the relative line ratios. We use olivine basalt composition listed in Näränen et al. [2] with total molar density of 0.13398 mol/cm^3. We pay special attention to the differences between the simulations and the FPE model, and extend the simulations to include the whole (θ_e,ϕ_e) hemisphere for different values of θ_i.

In Fig. 2 we show the relative differences between the simulated Fe Kα/ Ca Kα line ratios and the FPE-model for the olivine basalt material over the hemisphere for five r values, three ρ values, and $\theta_{\mathrm{i}} = 25°$. The main points are

- In the case of Fig. 2, increasing r and decreasing ρ lead to harder line ratios than expected from FPE modeling.
- Nevertheless, the behavior of the line ratios depend strongly on the incident spectrum. Especially, particulate media can also show softer line ratios than expected.
- The softening relative to FPE is observed for small ϕ_{e} and large θ_{e}. Increasing ϕ_{e} and decreasing θ_{e} harden the line ratios.

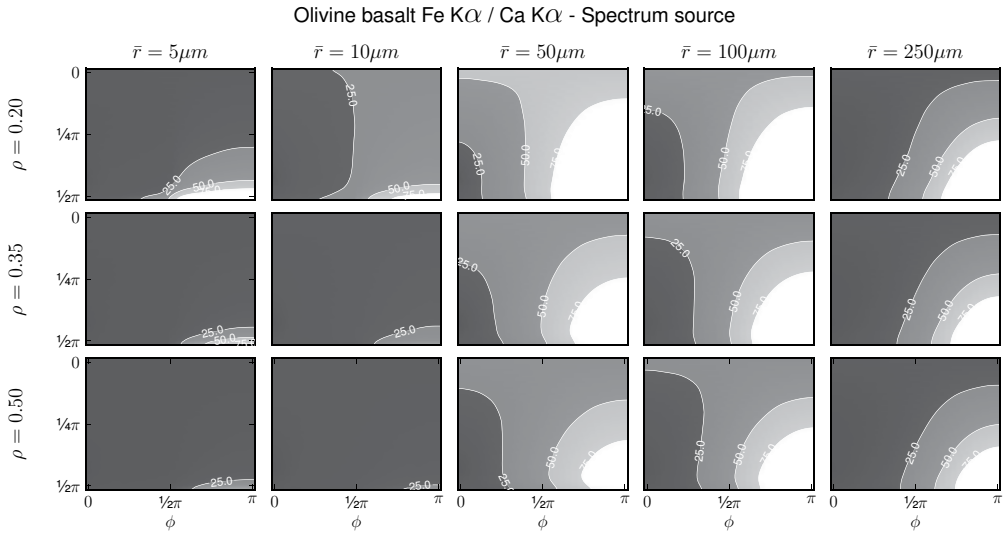

Figure 2. The relative difference between the simulated line ratios and the FPE-model. Dark gray corresponds to $< 25\%$ difference, white to $> 75\%$ difference.

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