

Accompanying text for the lecture on x-ray plasma spectroscopy

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1)

Hi, I'm Michal Šmíd (pronounced like Schmied :-), I will have this lecture on x-ray plasma spectroscopy. I find it more convenient for both me and you to provide you with a written script then to record the lecture the traditional way. So here you have this text which will guide you through the slides. You can just read this text while checking the figures in the presentation, the numbers here refers to slide number. I haven't copied the statements which are easily understandable from the bullet points in the slides, so it's better if you read those as well.

Currently I am a postdoc at HZDR, working in the group of Katja Falk. I'm working in the field of plasma spectroscopy since my bachelor thesis, which was already few years ago. I will be happy to get your questions or comments not only to this talk at my email address: **m.smid@hzdr.de**

2)

X-ray spectroscopy is one of many ways to study plasmas. What I like about it is that compared to most other ways, it is going directly to the ions, to their atomic structure and properties, thus giving you very deep information.

Far most of x-ray spectroscopy is studying line emission, i.e., the emission of radiative transitions between different atomic levels, therefore also majority of this talk is about emission line spectroscopy, but there is a wide area of other possibilities and effects which can be studied. Therefore, this talk does not try to be complete overview, but rather a brief look into the field with focus on topics where I have personal experience. In those topics I can give you rather deep and hopefully attractive look into what can be measured. More complete overview can be found e.g. in the book Atomic Physics in Hot Plasmas by David Salzmann.

Concerning the structure of the talk, it might be quite unusual. First I will introduce the so called Collisional-radiative model, which is an approach to simulate x-ray emission. Understanding of how this model will give you a good overview of the underlying physics.

Once you will understand how is the emission created, I will introduce the way how it can be measured, i.e. x-ray spectrometers. At the end, I will present four cases showing how spectroscopy can be used in 'real scientific life'.

3)

First, let's introduce the collisional-radiative model, which is an approach to computer simulations trying to reproduce the x-ray spectra. By going through the principle of such codes, we will nicely go through the theory.

If I should summary CR models in one paragraph, it would be like this: CR codes have a list of possible atomic states of ions present in the plasma, and a list of transitions between those states. The atomic states are static result of some other atomic calculation. The rate ('speed') of the transitions can be dependent on plasma conditions (mostly just density and temperature). So if you put those conditions as an input of CR code, then we can calculate the population of each of given

atomic state and also the amount how the transition happens. And since the spectra are determined by the transitions, then there is just a small step to get the synthetic spectrum. Then you compare this spectrum with what have you measured, and draw some conclusion.

Now look in detail at atomic states.

Each of such codes relies on more or less huge set of atomic data. By state we mean excitation and ionization configuration of ion, or in other words, atomic state mean how much electrons and in which orbitals an ion has. Here we see some examples of such states of aluminium or copper. So, the list of possible atomic states has to be an input of CR simulation, calculated by some atomic code.

Atomic state is basically a list stating which orbitals are occupied, a nice table reminding you of which orbitals exists is on https://en.wikipedia.org/wiki/Atomic_orbital#Orbitals_table.

The super-important variable, which is calculated by a CR code, is the fractional population of each state, " n_i ", denoting which fraction of ions is in given state " i ".

4)

Here we just visually illustrate an aluminum ion in four different states, so you can make sure you interpret the notation correctly. The left figure is neutral ion with 13 electrons, the other all various excitation levels of aluminum ion with two electrons (such is called He-like, because it looks like Helium - having 2 electrons).

5)

If we have the levels, we need transitions.

I.e. ways how ions can go from one state till another. These are described by rate coefficients, defining their 'speed'. The table and figure (from Salzmann) summarizes the basic processes and their inverse ones. You can see that while some processes are purely collisional (like electron collisional ionization), some involve radiation, i.e. absorption or emission of photon.

I recommend you taking a time to go through the picture and try to understand all those processes.

A silly quiz question: "Which process is the most important for x-ray spectroscopy?"

A silly answer: (think of it first yourself....) "It is the Spontaneous decay, because during this process, the transition lines in which we are mostly interested are produced." More correct answer would definitely be that all are important, because all can be responsible to set up the proper population. Neglecting any of those can lead to wrong results of the simulation. (More expert knowledge is to know, which are actually important and which could be in some situations neglected...)

6)

The set of atomic levels and atomic transitions makes up the 'atomic model', this can be pretty huge set of tables, often stored as ASCII files. Here you can see how it could look like for one popular code. The opened file is showing a fraction of transitions for 25-times ionized copper, the 2. till 5. column identifies to number of concerned state (i.e. the " i " and " j " in n_{ij}), the meaning of rest columns is dependent on the type of transition (defined by the first 'f' here). Typically it can be fitting coefficients of some formula providing the rate as a function of temperature (and density).

7)

If you now have the data, you have to solve it.

Actually you are solving those two equations. Actually, there is a small trouble. It is not two equations, the first one is a linear set of equations, as many as you have atomic states, which could be huge. Therefore some very specific algorithms need to be used to solve this set. The transition matrix W_{ij} contains sums of all atomic processes between levels i and j for given temperature and density. Luckily, due to the physics, this matrix is pretty close to be Block-diagonal matrix (https://en.wikipedia.org/wiki/Block_matrix#Block_diagonal_matrices), which significantly simplifies the solution, but can also bring some other trouble of numerical instability.

After solving the first equation(s), you could get the result:

time differentiation of population densities n_i . However it is still more complicated. Some processes contained in W_{ij} are dependent on radiation field I . And this is calculated by the second equation - the radiation transfer equation. The epsilon and kappa in this equation shows how radiation is created and absorbed in plasma with given atomic population. So to solve for I , you have to have n_i . As a result, you have to solve both equations together. And do not forget that the second equation is looking for $I(\omega)$, i.e. theoretically for a continuous function - radiation intensity at different frequencies ω . In practice, you discretize this function into reasonably small amount of energies.

Nevertheless, the model is still giving you just time derivative of population density....

8)

To know what to do with that we should look at plasma equilibria. I would assume you've heard of LTE and CE. And first let's look at an ideal case what we could do with CR model, so called **time dependent regime**. At the beginning of the experiment, we could assume the material is completely in basic non-excited non-ionized state, which could be $n_1=1$, and $n_i=0$ (for $i>1$). Also it would be on $T=0$, so there would be no radiation. (We have forgotten about some quantum effects now..) In this situation, you can solve the model and the result should be that your time derivatives are 0 and the system is staying in this state. Then you turn on some heating (preferably laser) - which means you would add another term into the second equation. The laser energy would drive ionization, excitation, increase of temperature, and the whole game would begin. This approach is however rarely used. First, because CR models might not be precise enough to predict temperature, second, because it is not needed and would be quite time consuming.

In processes which are not ultra-fast, you can use the assumption of **CR-steady state**. Which means that you assume that the population is not changing that fast compared to the changes of macroscopic plasma parameters (temperature..) Therefore you get the temperature from some other model or assumption, and you solve the equation $dn/dt = 0$. It is much easier, faster, but can break up sometimes.

See the two bottom pictures. It shows you the temporal evolution of **ionization distribution of a plasma** subjected to some heating. In both cases, the simulation used a temporal evolution of temperature from hydrodynamic simulation as an input. In the first figure, the CR-steady state model is used, i.e. for each time step I assume $dn/dt=0$ and I calculate the ionization distribution individually. From the figure you can see that the ionization (and temperature) have significantly risen at approx. 80-120 fs, and since about 150 fs, it is 'very slowly' falling down, so in the end still most of the population is in the He-like state.

The second figure shows the hybrid time-dependent regime. We are still using the temperatures from hydrodynamic simulation, but we assume that at the beginning, all ions were in ground state, and they are continuously evolving through the time. The beginning is similar - at about 150 fs,

most ions are fully ionized. And then a striking difference comes. The plasma stays almost fully ionized, although the temperature is going down same as in the previous figure. Why? For recombination, you need collisions between ions and electrons. But in this time, the plasma is already so diluted, that such collisions are rare - so it takes time until this happens. So to conclude - in this situation, the decrease of plasma temperature was faster then the atomic processes, therefore the CR-steady state is not valid....

This was just an illustration to help you get the feeling for the population equations. Don't worry much about it.

9)

This picture is trying to illustrate an extremely simplified CR model. Each bar represent one atomic level and horizontal levels most important transitions. Levels denoted by 'g' are the ground states, i.e. non-excited.

As Ly α is denoted the most prominent transition in H-like ion, that is ion with only one electron left. The transition goes between the 1s and 2p levels, i.e. going from the lowest excited state to the H-like ground state. Other arrows denote some other important transitions. The red & green bars shows the difference between LTE & CR-stedy state.

In LTE, the population is given by Saha equation, and it is assumed that collisions are significantly more important then radiation; the ionization is given by collisions.

In CR-steady state regime, the radiation plays a role in the system. This regime is actually a generalization of LTE, or in other words, both LTE and CE are special cases of CR-steady state for high and low density, respectively. The tax is that CR-steady state is much more difficult to calculate.

10)

Up till now we have calculated populations - but we need spectra! The four points shows how to easily get them; Each line is then supposed to have a shape given spectral line broadening effects described below.

11)

However, the fact that there exists a conference specifically aimed to discuss the spectral line shapes, shows us that it might not be that easy.

12)

Second issue is that the approach on previous slide will give you a spectrum at one region of plasma. As this spectrum is propagating through another parts of plasma, it can change. This is the spectroscopic radiation transport. For example, the peak of a spectral line can be absorbed more then its wings, therefore producing broadened line shape or even a line with a gap in the middle.

13)

Also, if the plasma is optically thin (like in the case of candle flame), the measurement will integrate along the ,line-of-sight'. And in the end we might be hardly sure from which plasma

element is the emission coming from. Typical approach can assume radial symmetry and use Abel inversion to handle that (https://en.wikipedia.org/wiki/Abel_transform)

14)

To further highlight the complexity of mentioned effects, there is yet another conference taking place every two years, where scientists are discussing various effects in plasma radiation. Particularly this only focused on 'Hot Dense Matter', i.e. not even all plasmas.

15)

And this conference is here just to compare various CR models. Because each model can produce different results, and we should know why and learn from each other.

16)

That was all now about the theory of CR models. As you have heard already about a lot of approaches for plasma simulations, I tried to make a super brief overview of most of them. The red text shows what can be simulated by given codes.

17)

xkcd.com

18)

This slide presents spectra calculated by a CR model, the top pane shows Aluminum plasma for various parameters. The difference between black and green spectra is only the plasma size - i.e. the increased intensity and line width is due to opacity. The red line is at higher temperature, therefore the relative contribution of Ly lines (produced in H-like ion) is larger compared to the He-like emission. The bottom pane shows similar situation at copper plasma. The wavelength range is scaled as Z^2 , and see that the lines are at approximately (but not exactly!) same positions. Also, the temperature is much higher in order to ionize copper to He & H-like states.

Note that here and in the rest of the talk I am using the units of Ångstrom, $1 \text{ \AA} = 0.1 \text{ nm} = 10^{-10} \text{ m}$. I'm sorry for this non-SI unit, but it was very common in the field of x-ray spectroscopy. Another very common unit for x-ray spectra is eV as an energy unit of photon, with conversion of

$$E [\text{eV}] = 12398 / \lambda [\text{\AA}]$$

19)

In this diagram I show a possible connection of various codes to produce synthetic spectra, which can be compared to experiments. On the top branch we define which atomic levels and processes are we interested in, and use some atomic code (Hullac in this case) to produce atomic data. The hydrodynamics is performed by combination of 2 codes to provide a 2D multi species data on the second line. Those goes together to the CR code Cretin, which calculates the desired spectra.

20)

Now we can quickly mention the detection possibilities. Actually I'll talk only about crystal spectrometers. The bottom picture shows example of Quartz crystal, which is a transparent glass-

like material inside a metal box (illuminated by expanded laser for alignment purposes). One experimental setup is shown in the right figure, where the red point is illuminated alignment needle, mimicking the plasma target. The crystal is in left part. The spectrum is detected on an x-ray film, which will be located in the black holder on the top. Currently, for alignment purposes, there is a white paper. We can see the red line in the center of the paper. This is made of the red light scattered from the needle, reflecting from the crystal and focused down to the paper.

Left figure shows x-ray films with recorded spectra - the dark lines. If possible, an x-ray camera or Imaging Plate is used instead of the film.

21)

Such spectrometers works on the principle of Bragg law. (https://en.wikipedia.org/wiki/Bragg%27s_law) That states that only those photons, whose wavelength fulfills the Bragg condition, are reflected. The condition is given by mutual interference of reflection from different crystal layers. If the curved crystal is used, the divergent emission can be focused and the spectrometer will provide a 1D spatial resolution, as is illustrated on the bottom figure.

There, an L-shaped 2 colored source is used as an an example. The radiation is reflected by spherically curved crystal. If the detector is positioned on the drawn circle, then radiation from points A and B is reflected to the same point in the dispersion direction (horizontal). Obviously, the different energies of photons (represented by red and green) are reflected to different positions. Due to the geometry, however, the radiation from point C is reflected to different line in vertical direction, providing the spatial resolution along this direction. This geometry can help separate radiation of the target surface from plasma emission form further regions.

22)

Let's have a look at the experiments now. There will be four different applications shown.

23)

The first one: plastic target was illuminated by laser. The plastic was doped by Chlorine, so the spectrometer was set up to see the Chlorine lines. The top picture shows the raw recorded spectra. Vertical direction is the spatial direction, horizontal spectral. The strong horizontal line is the emission directly from target surface. Higher is the emission further from the target, seen up till few millimeters. We have used $\text{Ly}\beta$ and $\text{He}\delta$ lines for the analysis, as these two are pretty well sensitive for temperature.

24)

A CR code called PrismSpect was used for this evaluation. By this code we have simulated single-cell steady-state spectra for parameters in the range $T= 500 - 950$ eV and densities between 0.02 and 0.5 g/cm³. Each of this synthetic spectrum was automatically fitted to the experimental data and their difference was calculated. The result is shown in the left figure. The agreement was best for $T=685$ eV and 0.1 g/cm³, so these values are used as the measured effective plasma parameters. Done.

25)

Another application is to use the famous Doppler shift to check the velocity of the plasma. Here we had a spectrometer observing the emission NOT exactly perpendicular to the laser axis. The plasma velocity in the axial direction is however so large, that even its projection to the small angle of the spectrometer observation (not shown in the figure) will produce measurable shift - in this case in the order of $1 \text{ m}\text{\AA}$ - and from this shift we can easily calculate the plasma velocity. The velocity was close to zero at the target surface, and was increasing as the plasma got further from the target. Additional obstacle was used in this experiment, the plasma hit the obstacle and the speed was getting down - as is seen in the Doppler shift close to the Mg foil.

26)

In different experiment with similar (but different) setup we could see another interesting effect. Now, the spectrometer was aligned exactly perpendicular, so the axial speed makes no effect. In the figure on the right (a), we can see a spectrum of two He α lines from just an Al target directly irradiated by laser. Interesting things happens when we, similarly as in previous case, introduce the obstacle. Then we can observe a splitting of both lines with an arrow-like shape.

By complex modeling, we could reproduce such shape by simulations (c). It is explained as caused by radial velocity of the reflected plasma, where the radial velocity was increasing with the increasing distance from the obstacle. Therefore the low energy and high energy wings of the lines corresponds to the plasma flying towards and away from the spectrometer. The central region of the plasma was actually too hot to emit those lines, so only the edges (surface) of the plasma cooled down enough to emit the lines.

27)

The third case is significantly more complicated, as the slides are based on one conference talk, it is introduced by this blank slide :-)

28)

The experiment was quite simple: Thin copper foil is irradiated by laser, and a spectrometer is observing the emission perpendicular to the target.

29)

This is how it looks in reality.

30)

And this picture based on hydro simulations is here to explain what can be happening. It is a snapshot of plasma density (blue) and temperature (red) close to the peak of the laser intensity. This laser has long enough duration (250 ps), so the first part of the laser already produced quite significant plasma. The peak of the density is the remnant of the foil, shifted by the laser about $20 \mu\text{m}$ left from its original position. There is a long plasma profile going against the laser up till $150 \mu\text{m}$, and even further. The laser is now absorbed at and below the critical density, which is around $100 \mu\text{m}$. Therefore this region has the highest temperature.

During the process of laser absorption, plasma instabilities takes place and produces so called supra-thermal electrons. Those can easily penetrate the whole target and get on its rear side. On their way, though, they collide with ions and can trigger collisional ionization in the K-shell. And this is followed by the emission of a $K\alpha$ line.

31)

Jumping to the other side of the problem, the red line is the measured spectrum. It is as we got it - without labels. Even to say which line is which needs quite a lot effort. In this case I have taken the atomic transition form the CR model and plotted them below on the same wavelength scale (blue points). The vertical axis is now the charge (aka ionization) state. All those lines are kind of the $K\alpha$ transition, that is 1s-2p transition. The wavelength is changing based on the ionization.

32)

Then I can easily draw vertical lines and label my experimental results by the name of the ionization state where it is originating. The right-hand figure is now illustrating how some of the emitting ions look like.

33)

To finish this, I add another plot to the right, now it is the dominant charge state for various temperature plasmas. Connecting the lines horizontally, I can see that for example, the line at 1.527 Å is originating in Fluorine-like copper, and this ionization state is dominant at temperature around 300 eV. The $He\alpha$, on the other side of the spectrum, is originating at around 2000 eV.

From this we can directly see that we cannot say at which temperature was this spectrum emitted. We can see that each line is emitted in different temperature, so the conclusion is that we are looking at plasmas with temperatures between 10 and 2000 eV. Which is correct. X-ray detectors are time and line-of sight integrated, so what we see is really integration over very different conditions.

34)

There are many different methods to untangle this, the choice (or rather design) of one is always dependent on what we want to learn from the experiment. In this case we wanted to know how much of those supra-thermal electrons are present in the target. Therefore how much of them was created in the interaction.

Se we have used a model of variation of plasma temperature and density as seen on the picture as an assumption, and we tried to find the amount of hot electrons (hot electron fraction) to reproduce the data.

35)

This means that our CR model had now 3 parameters: temperature, density, and hot electron density. We have sampled the temporal evolution in about 20 steps, and made a temporal integration of spectra over this range. As Temperature and Density are given, we have been varying only the hot electron density, namely those 20 values for each time step. The result is shown in red bars in

the figure. The black region is the margin of uncertainty. We see that for higher temperatures, the spectra is basically not sensitive to hot electrons. For temperatures below 400 eV, however, we have estimated the hot electron density as around $1e20 \text{ cm}^{-3}$.

36)

And here is the comparison of the best fitting spectra. For the complexity of the modeled phenomena, the result is quite impressive.

37)

Now we are at the last of the four presented cases. This time we are looking at absorption spectroscopy. In the experimental scheme below we can see the probe laser beam, which is converted into an x-ray beam on the gas target. This conversion is based on amazing Laser Wake Field Acceleration, but this is not important now. The orange color denotes the x-ray beam, which is going through the foil target (again thin copper foil), reflected and dispersed by the crystal and detected on the camera.

The spectrometer is observing the so called K-edge. The edge is defined by the energy needed to ionize an 1s electron from its bound state to continuum. Photons which have smaller than this energy cannot trigger this ionization, there are more likely to go through the target. Photons with higher energy ionizes matter by this process, therefore they are more likely to be absorbed. This produces a significant decrease in the transmission (the edge), as seen on the next slide.

38)

What is even more interesting for us is that the probability of absorption of photon above the edge is still dependent on its energy - there are small waves in the transmission spectrum.

The energy of the ejected electron is given by how much spare energy the photon have after the ionization. Therefore for photons only slightly above the K edge, the electron have very small residual energy. Such ,slow' electron is strongly interacting with the neighboring ion, and can undergo either positive or negative interference. This in turn modifies the absorption cross-section for given energy.

Long story short: the ,waves' above K edge are corresponding to the ionic structure of copper. If we see them as in the bottom picture, the copper is in solid crystalline structure. If it gets melted, they disappear and another ('liquid') peak will appear. Since this diagnostic can be (compared to emission spectroscopy) time resolved, we can study the ultrafast process of melting and creation of Warm Dense Matter.

38)

That is all from me now, thank you for your interest, and again, any questions or comments are welcome on my email address.