Warm Dense Matter and Strongly Coupled Plasmas Created by Intense Heavy Ion Beams and XUV-Free Electron Laser: An Overview of Spectroscopic Methods

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Abstract:
High density plasma physics, radiation emission/scattering and related atomic physics, spectroscopy and diagnostics are going to make large steps forward due to new experimental facilities providing beams of intense heavy ions and X/XUV free electron laser radiation. These facilities are currently being established at GSI-Darmstadt and DESY-Hamburg in Germany to access new and complementary parameter regimes for basic research which have never been obtained in laboratories so far: homogenous benchmark samples near solid density and temperatures from eV up to keV. This will provide important impact to many disciplines like astrophysics, atomic physics in dense environments, dense and strongly coupled plasma effects, radiation emission, equation of state. The spectroscopic analysis of the radiation emission plays a key role in this research to investigate the dynamics of electric fields in multi-particle coupled Coulomb systems and the modification of plasma statistics.

Key-Words: spectroscopy, diagnostics, highly charged ions, dielectronic satellites, x-ray scattering, non-equilibrium plasmas, laser produced plasmas, Warm Dense Matter, strongly coupled plasmas, heavy ion beams, free-electron laser

I. Introduction
Dense strongly coupled plasmas (DSCP) and Warm Dense Matter (WDM) refer to high-density-plasma regimes where atoms and ions behave in a manner intrinsically coupled to the plasma itself: the plasma exhibits long- and short-range order due to the correlating effects of the atoms and ions. This intriguing regime where the plasma environment can no longer be considered a thermal bath and the behavior of its particles is no longer well described by characteristics of isolated ions provides a tremendous challenge to researchers.

The relevance of DSCP/WDM research arises from a wide occurrence of this state of the matter: it is a subject of investigation in planetary science, cold star physics and all plasma-production devices where the plasma generation starts from cold dense matter (e.g., laser solid matter interaction, heavy ion beam driven plasmas, capillaries, exploding wires, and pinch plasmas). The character of interactions among the plasma particles is defined by the
ion-ion coupling parameter $\Gamma_{ii}$, i.e., the ratio of their inter-atomic potential energy to the kinetic energy given by

$$\Gamma_{ii} = \frac{1}{4\pi\varepsilon_0} \frac{Z^2 e^2}{kT} \frac{1}{R} = 2.32 \cdot 10^{-7} \frac{Z^2 n_{ii}^{1/3} \text{(cm}^{-3})}{T_i(\text{eV})},$$

(1)

$Z$ is the ion charge, $T_i$ the ion temperature and $R$ is the inter-particle spacing. The most intensively studied plasma regimes span the density-temperature phase space from modestly coupled ($\Gamma < 1$) to strongly coupled ($\Gamma > 1$), Figure 1 shows the diagram for hydrogen.

Figure 1: Density-temperature phase space diagram of hydrogen, WDM = Warm Dense Matter, HDM = High Temperature Density Matter, the coupling parameter $\Gamma = 1$ is indicated separating strongly coupled ($\Gamma > 1$) and weakly coupled plasmas ($\Gamma < 1$).

The current knowledge of the plasma behavior is particularly limited in those regions where standard theoretical approaches fail and experiments are exceedingly difficult. What concerns the DSCP/WDM, the difficulties in the field of theory arise from the lack of obvious expansion parameters, as the usual perturbative approaches used in plasma phase theories are no longer valid. Further, the importance of density effects (e.g., pressure ionization) increases, as the plasma environment starts to influence substantially the internal structure of ions or atoms. Experimental problems follow from a difficult isolation of well-defined plasma samples and their complicated diagnostics. This is why we know so little about it [1].

The current state-of-the-art indicates that the hitherto experimental conditions for generation of strongly coupled dense plasmas have been far from the desired optimum:

a) optical lasers can access the high temperature regime (some 100 eV), but they do have problems with sample homogeneity and also with the temperature scaling (the higher the irradiating intensity $I\lambda^2$, the more disturbing hot electrons are generated; these hot electrons seriously influence the plasma parameters, thus the near solid density samples are difficult to prepare and to analyze). Already the VUV-Free Electron laser will allow to overcome these essential drawbacks, because the $I\lambda^2$-parameter is in the classical regime due to the short laser wavelengths.

b) High intensity ions beams can overcome the drawbacks of laser produced plasma experiments (essentially inhomogeneity and short time scales), however, only temperatures up to several 10 eV can be reached even with the most powerful beams of future accelerators (FAIR at GSI-Darmstadt).
For these reasons, intense heavy ion beams and X/XUV/VUV free electron laser radiation are complementary.

The analysis of the radiation emission/scattering originating from the various elements and ionization stages is of great interest because it may provide the possibility for a unique characterization of the plasma [e.g., 2-12]. A large impact of the spectroscopic analysis stems from the fact that it is based essentially on a collisional radiative approach and therefore provides an information on plasma conditions independent of plasma models. However, standard spectroscopic methods are invalid because the environment of dense strongly coupled plasmas leads to an essential modification of atomic spectra and atomic processes [3, 13]. The plasma ion microfield results in a strong "mixing" of atomic energy states which requires to go beyond the standard consideration, namely to the density matrix representation. The second step is the frequency fluctuation method [14] to describe the "mixed" atom representation where all transition probabilities contain the effect of plasma electric field. These approaches will allow to take into account the effects of dense plasma environment on atomic processes [13].

Therefore new developments are urgently requested which, on the other hand, provides also a great challenge to the researchers: revision of the line intensity distribution (different from standard Stark theory), revision of the radiation trapping, revision of the interrelation between discrete and continuum spectra, decrease of ionization potentials, cut off of the statistical sums, modification of the three-body recombination in electric field, modification of the Saha-equilibrium relations. These effects will dramatically influence on the properties of the media: ionization balance, radiation loss, charge state distribution, equation of state. The development of new spectroscopic methods to analyze the radiation emission is therefore strongly related to the basic research of dense strongly coupled plasmas.

II. Solid density plasmas created by VUV-FEL irradiation of solids

The free electron VUV/XUV-laser (FEL) at DESY (Germany, operation in 2005) will allow to create samples near solid density at several 100 eV. Due to the small laser wavelength, the radiation can deposit energy directly into the volume (complicated surface effects which exist for optical laser are therefore avoided) because the laser wavelengths corresponds to the plasma frequency $\omega_p$ of a solid density plasma:

$$\hbar \omega_p = 11.2 \text{ eV} \sqrt{\frac{n_e}{10^{23} \text{ cm}^{-3}}}.$$  \hspace{1cm} (2)

For fully ionized aluminum $\hbar \omega_p = 33 \text{ eV}$ which corresponds to the energy of the photons generated by the FEL. Therefore, interesting resonance phenomena can be studied. As the laser pulse duration is only about 100 fs, the sample will be heated before essential expansion can take place.

Moreover, the hot electron parameter $I_{\lambda}^2$ (the larger the parameter, the larger the hot electron production) is likewise small and the plasma creation is expected in the classical regime without hot electrons. In contrast optical lasers do have the disadvantage to create hot electrons at high intensities and therefore do not permit to compare samples of different temperatures. Therefore, for the first time near solid density benchmark samples can be created with the VUV-FEL in a wide temperature interval (50 eV until keV). The provoked self x-ray emission can be used for advanced diagnostics.

In the end of 2005 the VUV-FEL will provide 50-100 $\mu$J pulse energy at 30 nm wavelength, a focus of 2 $\mu$m and a pulse duration of 20 – 50 fs. These parameters allow to perform soft x-ray spectroscopy (K-shell emission) of the self emission spectra by means of curved Bragg crystals (two-dimensionally curved x-ray crystals provide simultaneously high spectral and spatial resolution while maintaining high luminosity: no slit). The number of
photons can be estimated as follows: the laser volume (2 \mu m \times 2 \mu m \times 100 \text{ nm} target thickness) contains \(2.4 \times 10^{10}\) Al particles. If we assume that only 20 \% of the 100 \mu J energy is transferred to the particles, a thermal energy of 5200 eV is available for each particle which is sufficient to ionize all particles until the H-like charge state. Assuming a CCD efficiency of 20 \%, second order mica crystal reflectivity for He\(\alpha\) of 1 \%, collection of solid angle by means of spherical crystals of 0.1 \% and assuming that only 1 \% of the radiation emission is going into the He\(\alpha\) radiation, we arrive at about \(4 \times 10^4\) He\(\alpha\) photons per shot. For He\(\beta\) and satellite transitions we expect about \(10^3\text{-}10^4\) photons. This is enough to obtain a single shot K-shell spectrum with good signal to noise ratio. The signal to noise ratio can be further improved making use of the 10 Hz repetition frequency and rotating targets.

Figure 2: Parameter evolution of VUV-FEL irradiated solid Al foil, thickness \(d = 100\) nm, laser wavelength \(\lambda = 60\) nm, pulse lengths \(\tau = 100\) fs, \(I = 10^{16}\) W/cm\(^2\). The MULTI-fs simulations starts with \(Z_{\text{min}} = 4\) and the target is divided into 120 cells. Laser propagation starts from cell 1 to cell 120.

Figure 3: MARIA simulations of the time evolution of the ionic abundances of a Mg target heated by VUV-FEL. H = H-like Mg, He = He-like Mg etc.

Figure 4: MARIA simulations of the time evolution of different resonance and satellite lines. Line opacity is taken into account.
Using the time evolution of the parameters shown in Figures 2 we have calculated the x-ray emission of an Mg-target with the MARIA-code [7]. Figure 3 shows the evolution of the ionic charge states (H=H-like, He=He-like Mg etc.). At the beginning of the laser pulse the plasma rapidly ionizes until the He-like charge state. After the laser pulse is switched off, recombination processes led again to a strong population of the He-like, Li-like and lower charge states (note, that the constant fraction of the Be-like ions for longer times is an artefact: the hydro-simulation was stopped at a certain lower temperature and the spectroscopic system was allowed to relax even after that time).

The recombination processes cause intensive radiation even after the laser pulse is switched off. Figure 4 shows the temporal behavior of $H_{\beta} = 1s^2 - 1s2p \; ^1P_1$, $L_{\alpha} = 1s - 2p \; ^2P_{1/2,3/2}$, Li-like satellites $1s^22l - 1s2l2l'$ and Be-like satellites $1s^22s^22p^m - 1s2s^2p^m$ with $n+m=3$. It can be clearly seen, that long lasting strong emission of $H_{\beta}$ and even the Li-like satellites takes place. The long lasting emission (ps-scale) at rather low densities (see Fig. 2) overlaps with a short heating phase (100 fs scale) and therefore will mask the high density signatures in the spectrum. It should be noted that no streak camera is available for a 50 fs-time scale in order to extract only the heating phase.

We are therefore left with the general problem to look for “Fast X-ray Emission Switches”, i.e. line emission which is intensive only in the heating phase. The simulations show, that the general class of resonance lines is not suitable, because they can be strongly populated by radiative recombination which has no low energy threshold for highly charged ions. Dielectronic satellite emission can serve for fast x-ray switches as can be seen from the time evolution of the Be-like satellite emission, Figure 4. It is very remarkable that the emission time of the Be-like satellites is only about 100 fs although the population of the Be-like ions remains strong for several ps. This effect is related with the resonance effect of the dielectronic capture processes. Only those free electrons are captured from the continuum, whose energy matches the so called dielectronic capture energy $E_C$. For highly charged ions, the capture energy is so large, that at low temperatures (the recombination regime) the emission is practically exponentially cut off.

$$\Gamma \approx \alpha \cdot n_e \cdot \frac{B \cdot g_s \cdot \exp(-E_C/T_e)}{g_k \cdot T_e^{3/2}}.$$  \hspace{1cm} (3)

$T_e$ is the electron temperature in [eV], $B$ is the branching factor, $g_s$ and $g_k$ are the statistical weights of the upper and lower levels, $\Gamma$ the autoionising rate in [s$^{-1}$], $n_e$ the free electrons density [cm$^{-3}$], $n_k$ the population density of the level into which dielectronic capture takes place and $\alpha = 1.658 \cdot 10^{-22}$ cm$^3$ s$^{-1}$.

Figure 3 indicates, when using, e.g., B- and C-like dielectronic satellite transitions, a time scale of the order of 10 fs can be obtained because exponential cut off and short life time of lower charge states are both in favour for a fast x-ray emission switch [17].

Table 1 shows the electron temperature in dependence of the the VUV-FEL intensity. It can be clearly seen, that between the intensity range of about $10^{15} - 10^{17}$ W/cm$^2$ leads to a very attractive temperature range between some 10 eV and 1 keV which can provoke K-shell line emission of, e.g., Mg, Al.

MULTI-fs simulations [18] of the electron temperature of a $d = 45$ nm thick Al target, photon energy $E_\omega = 30$ eV, pulse shape : $\sin^2$ pulse, $\tau = 100$ fs

| $I$ [W/cm$^2$] | $kT_e$ [eV] |
|---------------|-------------|
| $10^{14}$     | $\approx$ 1 |
| $10^{15}$     | $\approx$ 20 |
| $10^{16}$     | $\approx$ 300 |
| $10^{17}$     | $\approx$ 1300 |
| $10^{18}$     | $\approx$ 4000 |
III. Warm Dense Matter created by intense heavy ions beams

Making samples of WDM suitable for study in the laboratory is difficult. Laser-plasma generated samples tend to suffer from steep density and temperature gradients and are short lived (~ns). The ion beam facility at GSI offers the chance to make large uniform samples with scale times of ~100 ns: the sample size is mm, homogeneity is achieved by positioning the Bragg peak outside the sample and the long live time is essentially given by the hydrodynamic time scale of the large sample.

However, heavy ion beams will provide only temperatures in the eV range (several 10 eV are envisaged: GSI FAIR project). At so low temperatures, x-ray self emission cannot be provoked.

Therefore x-ray scattering driven by the kilojoule PHELIX laser has been proposed for diagnostic purposes [19]. The scheme of the x-ray scattering diagnostic is depicted in Figure 5. Spectrally resolved X-ray scattering is supposed to provide information about the free electron density, the electron temperature, the chemical composition and the plasma coupling parameter. Theoretical work on X-ray Thomson scatter spectra, looking at the effects of strong coupling and degeneracy has been started [20] and is expected to be stimulated further by the availability of data from interesting samples such as dense warm hydrogen.

Because the scattering cross sections are rather low, a high laser beam energy of several kilojoule energy is requested. We intend to use the ~ 1 kJ available PHELIX-beam to generate ~ 10^15 photons [21] of Ti He-alpha (1s^2-1s2p ^1P) line radiation at 4.75 keV. We can consider how many X-rays are likely to be scattered. The X-ray source sample (Ti foil) can be placed within 5 mm of the sample. A pinhole system can be used to restrict the probe to a fixed diameter of the target. Assuming this is done and the probe subtends ~0.3 mm on the target we can estimate that ~10^{12} photons probe the target. The scatter cross-section depends on the scatter angle chosen but is ~4·10^{-26} cm^2/sr. As an aside, we should state that the angle of scatter determines if the scatter is in the collective or non-collective mode. In the former we sample fluctuations on a longer spatial scale than the Debye length and expect to see a plasmon feature in the scatter spectrum, in the latter we probe shorter spatial scales and expect to see a spectrum reflecting the Doppler shifts due to thermal motion of the electrons. Give the size of the plasma we expect to see at least 10^8 photons/sr scattered. With a spatially resolving imaging crystal we can achieve a throughput of 10^{-4} sr and thus capture 10^7 photons per data shot. Given the high detection efficiency of modern CCD X-ray detectors, this is enough to make a spectrum. The expected spectra should differ from optical spectra in that the Compton shift of scattered photons (0-90eV) is noticeable and the spectrum is not symmetric since the structure factors on either side of the central wavelength are related by S(-\omega,k)~e^{-\omega/kT}S(\omega,k). This is not important, in the optical case (\hbar\omega<<kT) but is for X-rays and collective scatter would yield a single plasmon peak shifted by the plasma frequency plus Compton shift from the incident wavelength.

Figure 5: Scheme of the experimental scattering diagnostic driven by the kilojoule PHELIX laser.
Figure 6: Static confinement scheme leading to homogenous distribution of temperature and density. The massive stiff shell is a great disadvantage as it does not permit the application of scattering diagnostics due to the large photo-absorption even on µm-scale.

The final stage of interest in view of the future SIS-100 accelerator construction would be to shorten the pulse length down to some 10 ns and increase the number of particles to several $10^{12}$ in order to heat bare samples under almost homogenous parameter conditions to several eV (the pulse lengths is sufficiently short that the sample has no time for serious expansion resulting in inhomogeneities). However, these parameters will be achieved only in the last step of the accelerator construction.

In view of this long lasting period of almost 10 more years for the final completion of the GSI future accelerator concept, highlight experiments and new concepts in the intermediate construction stage of the future GSI facility are highly requested: to keep the researchers motivated on the long way of construction and to support the future accelerator concept with scientific excellence.

For these purposes, we propose a particular experimental arrangement for the Warm Dense Matter research of solid hydrogen for future SIS-18 upgrade parameters. The method is based on a dynamic confinement scheme rather than a static one. Figure 6 (left) shows the well known static scheme which leads to homogenous samples of Warm Dense Matter, see parameter evolution in Figure 6 (right) [22]. The achievement of homogeneity is simply based on the fact that the massive shell is not moving due to its inertia and therefore confines the internal hydrogen. This scheme, however, has a serious disadvantage: the photo-absorption in the outer confining shell (e.g. lead or tungsten) is so large that no X-ray photons produced with the PHELIX kJ-pulse for x-ray scattering purposes will reach the sample.

Figure 7 shows the proposed dynamic confinement scheme [12]. The outer shell of a low Z-element is heated along with the sample thereby providing a pressure towards the axis and allowing X-ray scattering diagnostic through the low-Z-tamper.

The scheme can be optimised in an impressive manner. Taking Carbon phenolic with $\rho_C = 1.5 \text{ g/cm}^3$ (70% carbon, 30% phenolic resin) as a tamper, $R_H = 300 \mu\text{m}$, $\Delta R_C = 50 \mu\text{m}$, and the envisaged SIS-18 upgrade parameters almost static parameter conditions are met [12], see Figure 8.
Figure 7: Scheme of the experimental arrangement of a cryogenic tampered sample heated by heavy ion beam. The plasma is diagnosed by x-ray scattering driven by an high energy laser.

Figure 8 shows that the dynamic confinement scheme is rather close to the isochoric heating which is the ideal case. By variation of the tamper thickness the target can be optimised to yield exactly the initial hydrogen density at the end of the heating beam bunch or to minimize the fluctuation of the hydrogen density during the whole heating time.

Concerning SIS-100, the further advantage of the dynamical confinement scheme is that excellent research in the Warm Dense Matter can be done making use of any progress in the beam intensity and pulse shortening. Therefore, all intermediate steps of beam intensity and pulse length improvement can be translated into valuable experiments addressing continuously new parameter regimes of Warm Dense Matter.

Figure 9 (left) shows the density evolution of a bare hydrogen sample under the parameter conditions envisaged for the SIS-100 accelerator. Due to the short pulse length, even the bare sample has rather homogenous density evolution, and maximum temperatures
of about 2 eV are achieved. These are outstanding conditions: Solid density, strong self emission (optical range, e.g., Hα,β) due to sufficient temperature, a rather high degree of ionization to create strong electric micro fields and the homogeneity will allow X-ray scattering from the whole sample to make interpretation of the data simple.

Figure 9: Evolution of the density of bare samples of cryogenic hydrogen/neon assuming SIS-100 parameters (pulse length 30 ns, 10¹² particles U²⁸⁺, energy 2.7 GeV/nuc).

Figure 9 (right) shows the parameter evolution of a cryogenic neon sample heated by an ion beam envisaged for the SIS-100. Temperatures of up to 7 eV will be reached and this will lead even to considerable electron impact induced ionisation. We therefore expect not only electron micro fields to play a decisive role but ion micro fields too. Transitions of interest in the optical range would be 3p – 3s transitions of, e.g. Ne I and Ne II: The lower states are excited states which will lead to a strong reduction of the bound-bound transition opacity. We note that the possibility to observe the 3p-3s transitions has already been demonstrated [23].

We note, that heavy ions have typically energies of some 100 MeV/nuc and therefore inner-shell x-ray transitions (Kα and Kβ transitions) from targets with nuclear charge Zₐ > 10 can be excited. At present, however, it is unclear how their spectral distribution can be interpreted to investigate plasma parameter dependencies for diagnostic purposes.

As the density is near solid density, predictions of the free electron density, Stark broadening, and continuum lowering are almost impossible and just the experimental observation of the existence of several line transitions will provide an important impact to the theory. Moreover, the additional scattering diagnostic will permit to verify and stimulate theoretical development on a quantitative level as the homogeneity of the mm-size samples is outstanding and the energy input to the sample by means of heavy ion beams can be measured very well.

We also would like to mention a recently proposed effect concerning the scattering of radiation induced by the heavy ions itself [24, 25]. The scattering of the electromagnetic field by highly charged ions in a dense medium might be useful for the WDM structure determination. As the effect is proportional to Zₐ² of the projectile charge and increases strongly when attaining relativistic velocities we do met favorable conditions for the scattering in the case of intense heavy ion beams.
IV. Conclusion

Intense free electron x-ray laser radiation and intense heavy particle beams will allow to produce plasma samples in high density parameter regimes never obtained in laboratories so far. Each of these methods has particular experimental advantages for the sample production which are both essential. The parameter regimes are complementary and differ essentially in temperature (eV up to 10 eV for intense heavy ion beams, up to keV for the FEL radiation). Novel x-ray spectroscopic techniques (curved Bragg crystals, x-ray scattering, higher order satellite transitions, density matrix approach) play a key role in the research to investigate dynamics of electric fields in multi-particle coupled Coulomb systems and the modification of the plasma statistics, i.e. strongly coupled plasma phenomena and Warm Dense Matter where we do know only little about:

1. Transition from discrete to continuous spectra, ionization of atomic states in plasma micro-fields and effects of strong coupling on the transition from discrete and continuous spectra.
2. Modification of atomic kinetic models in dense plasmas: “Mixing” of atomic energy states by plasma microfield, density matrix method and FFM method for calculations of spectral line shapes and population kinetics.
3. Spectral line shifts due to the dense strongly coupled plasma environment and consideration of plasma screening effects, collision induced shifts and polarization shift. Line positions are most significantly altered in plasmas where the potential energy is comparable to the kinetic energy of the particles, i.e., at large Coulomb coupling parameters $\Gamma$. High spectral resolution data will therefore provide the necessary basis for the comparison with theories (line shifts and line broadening – controversial discussion between various theories, continuum shifts, level depression).
4. Modifications of autoionizing rates, dielectronic satellites and spectral line intensities due to plasma micro-fields.

This will provide important impact to many disciplines like astrophysics, atomic physics in dense environments, dense and strongly coupled plasma effects, radiation emission, equation of state.

The particular advantages of the spectroscopic approaches are the independent (independent from plasma models) characterization of the unknown samples. This will have a large impact to the wide field of strongly coupled plasma theory. The spectroscopic methods and simulations to be developed will be of general interest for the dense plasma physics and atomic physics (in complex environments) community and stimulate further developments in x-ray spectroscopy and the simulations of the radiative properties.

In traditional atomic physics (of isolated atoms and ions) a very high accuracy has nowadays been reached. In contrast, the uncertainty in atomic physics in dense complex environments might be orders of magnitude. Therefore, principle effects and mechanisms are not yet understood and independent spectroscopic methods play an exceptionally important role in the research.

V. References

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