Integrated Analysis of Spectroscopic Data

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Spectroscopic data are analysed by fitting a collisional-radiative model to the emission spectrum of a low-temperature plasma in the wavelength range of visible light. The inference procedure employs Bayesian probability theory and accounts for all measurement and model uncertainties. This allows for the validation of model parameters, such as atomic data obtained in recent close-coupling calculations, which are ore only partly or not at all accessible by beam-type experiments. Initial results indicate that the spectroscopic data contain significant information about some Einstein coefficients for spontaneous emission of less prominent spectral lines.

Keywords: Integrated Data Analysis, Optical Emission Spectroscopy, Neon, Edge Plasma, Atomic Data

1 Introduction

The line radiation emitted by excited neutrals in a plasma can be used to obtain information about characteristic plasma parameters. More specifically, properties of the plasma species causing the excitation can be assessed. The energy dependence of the elementary processes allows, to detect deviations from thermal energy distribution functions. In the case of low-temperature plasmas, the main excitation channel is electron impact excitation, and thus the spectrum carries information about the electronic component of the plasma.

An established technique is to analyse the relative magnitude of line intensities measured, for example, in neutral-beam plasma diagnostics to infer the electron density n_e and the temperature T_e [1]. The approach presented here uses a collisional-radiative model (CRM) to reconstruct electron energy distribution functions from a whole range of the spectrum of a cylindrical neon discharge plasma. The population of different atomic levels is described by a set of balance equations. With these level populations, the light emission along a line of sight can be modelled. Hence, starting from the electron energy distribution function (EEDF), these steps lead to a full forward model of the spectroscopic data, incorporating the intensities of 87 emission lines. The approach described in [2] is extended by a description of the full spectral data. The basic idea is to calculate the probability of fitting the forward model to a full spectrum. The inference is done in the framework of Bayesian probability theory.

For the data analysis approach in Bayesian probability theory, it is necessary to describe all uncertainties involved. In addition to error statistics of the data, the uncertainties of model parameters are incorporated in the analysis. For the interpretation of spectroscopic data, Einstein coefficients and excitation cross sections are such model parameters. The provision of a complete and validated data set is a persistent issue in the interpretation of any spectroscopic measurement. The incorporation of the uncertainties of the extensive sets of atomic data is a challenging issue, because it can involve the probabilistic description of a great number of parameters. Since resolving this issue is a prerequisite for any uncertainty assessment in the interpretation of such spectroscopic data, the validity of the employed atomic dataset with respect to the spectroscopic results is addressed below.

A particular benefit of the approach presented in this paper results from the use of a full collisional-radiative model, which accounts for all correlations in the data, i.e., how the line intensities are affected by all transitions relevant to the observed spectrum. This enables using the spectral lines to assess the validity of the atomic data used for the collisional-radiative modelling.

The spectroscopic data to be analysed are obtained from a cylindrical neon discharge, a well-investigated system with various published results (e.g. [6, 7]) that can be used for validation. The full set of atomic data being employed was calculated by a *B*-spline Breit-Pauli *R*-matrix (close-coupling) model, as described in detail in refs. [3, 4].

2 Data descriptive model of the spectroscopic measurement

The forward model maps the quantity of interest, the EEDF $f_e(E)$ onto a simulation of the measured data $\vec{D}$ (spectrometer pixels). It consists of a chain of different elements described below. More details about the data model can be found in [5]. Schematically, we have

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$$\underbrace{\underbrace{f_e(E)}_{\text{kinetic theory}} \rightarrow n_i}_{\text{collisional radiative model}} \rightarrow I_{ij} \rightarrow \underbrace{\int_{\text{l.o.s.}} I_{ij} dV}_{\text{radiation transport}} \rightarrow L(\lambda) \quad \underbrace{\rightarrow \vec{D}}_{\text{measurement}} .$$

Different parameterisations of the energy distribution are employed to determine electron collision rates for the collisional-radiative model (CRM). Here, the data are well described with a Druyvestein distribution. EEDFs derived from hybrid modelling of neon discharges accounting for a kinetic treatment of the electrons [6, 7] are used to validate the obtained result. The CRM consists of a set of balance equations for the population densities n_i of 31 excited states of neutral neon, taking into account populating and depopulating elementary processes.

The *locally emitted power* $I_{ij}\left[\mathrm{W}/(\mathrm{m}^3 \cdot \mathrm{sr})\right]$ can be readily obtained by multiplication with the inverse lifetime of the excited states and the photon energy, and division by the full solid angle (4π).

The radiation has to pass through the plasma before it leaves the discharge device. The apparent lifetime of the excited states is affected by the transport of photons if the plasma is optically thick, i.e., for transitions to the ground or metastable states of the atom [8]. The description of this opacity, together with the integration along the line of sight (l.o.s.) of the spectrometer, yields the *effective spectral radiance* $L(\lambda)$ as a function of the wavelength λ.

The modelling of the actual measurement comprises the translation of $L(\lambda)$ into the detected signals and the mapping of wavelengths to pixel numbers. This requires details on the detector response, which were measured with a standard light source (sensitivity calibration). The wavelength mapping is fitted to the data within the reconstruction.

Figure 1 shows the result of the forward model together with the measurement. The parameters giving the best fit are obtained by maximising the probability distribution function (PDF) for the parameter vector $\vec{\Theta}$, which is called the *posterior* $P(\vec{\Theta}|\vec{D}, I)$. It describes the probability of a certain parameter set to be true, given the measurement $\vec{D}$. The posterior is set up according to Bayes' rule:

$$P(\vec{\Theta}|\vec{D}, I) = P(\vec{D}|\vec{\Theta}, I) \cdot \frac{P(\vec{\Theta})}{P(\vec{D})} \tag{1}$$

It contains the *likelihood* $P(\vec{D}|\vec{\Theta}, I)$ and the *prior* $P(\vec{\Theta})$. The *evidence* $P(\vec{D})$ is not taken into account, since it does not affect the position maximum of the posterior and can be deduced from the normalisation constraint. The likelihood corresponds to quantifying the probability of a certain outcome of the measurement, given the true and unknown state of the system $\vec{\Theta}$. For a Gaussian distribution of the error statistic of the spectrometer pixels, it is given by

$$P(\vec{D}|\vec{\Theta}, I) = \frac{1}{\prod_i \sigma_i (2\pi)^n} \exp\left\{-\frac{1}{2} \sum_i \cdot \frac{(D_i - D_{\mathrm{sim},i})^2}{\sigma_i^2}\right\} \tag{2}$$

The prior is describing the knowledge about the model parameters which is not contained in the data, but originates from independent sources. For example, atomic data that are subject to uncertainty may be described by means of a parameter with a prior distribution reflecting the known confidence region.

The marginalisation theorem for probability distributions describes how to integrate out parameters $\vec{\eta}$ we are not interested in:

$$P(\vec{D}|\vec{\Theta}, I) = \int P(\vec{D}|\vec{\Theta}, \vec{\eta}, I) d\vec{\eta}. \tag{3}$$

In the data model there is no formal distinction between the so-called *nuisance parameters* $\vec{\eta}$ and the *parameters of interest* $\vec{\Theta}$. The result of the projection down to the dimensions of the parameters of interest is a broadened PDF compared to a posterior not taking into account additional parameters from the beginning. The parameters of interest in this analysis are either the EEDF or Einstein coefficients we are interested in for validation purposes. A table with all relevant nuisance parameters can be found in [9].

3 Atomic Data for Collisional-Radiative Models

In the forward model a consistent set of atomic data, obtained from a *B*-spline Breit-Pauli *R*-matrix (BSRM) model for the treatment of e−Ne collisions [3], was taken as the starting point. The BSRM approach is based on the close-coupling approach. In contrast to all other commonly used perturbative or non-perturbative methods to generate such atomic data, the use of non-orthogonal, term-dependent sets of atomic orbitals makes it possible to obtain fairly accurate descriptions of both the energy levels and the oscillator strengths with comparatively small configuration-interaction expansions. In traditional methods with orthogonal sets of one-electron orbitals, a similar accuracy can, in principle, be achieved by very large expansions using so-called pseudo-orbitals [3].

The major advantage of using this dataset is the full provision of all required data for the collisional-radiative model, which goes far beyond the availability in standard databases such as [10]. Furthermore, the wavefunctions for the basic states are derived consistently from common structure calculations. This means that all derived atomic data, such as oscillator strengths and cross sections, depend on the same set of wavefunctions. From a detailed analysis of the calculations, critical matrix elements can

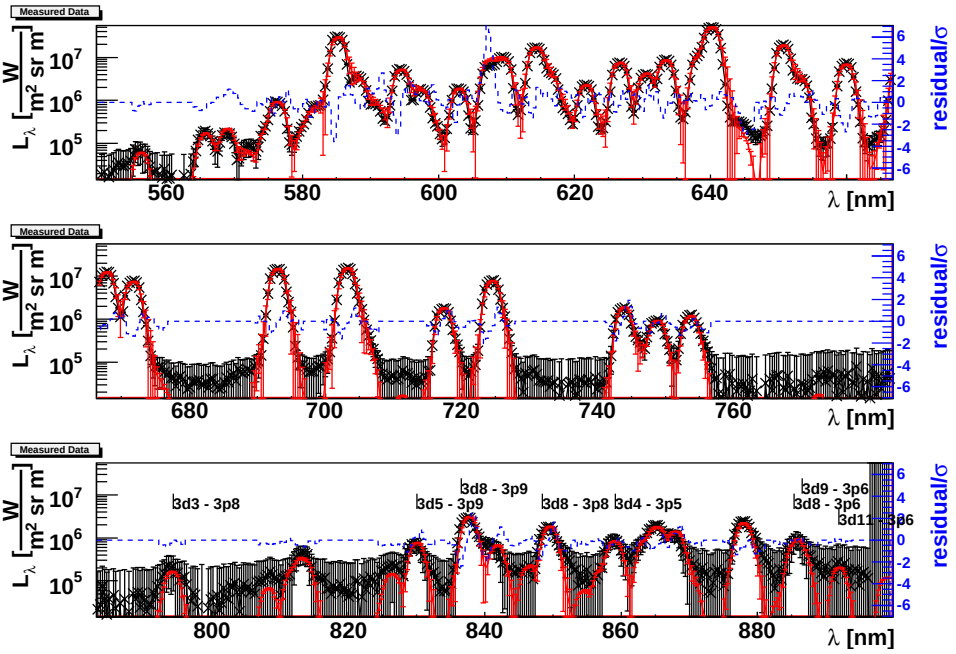

Fig. 1 Result of the forward model. The red line depicts the modelled spectrum, black crosses show the measurement and its uncertainties, and the dashed line is the difference between the two in units of standard deviations. The labels in the lower part of the spectrum mark lines whose Einstein coefficients are extracted from the spectrum.

be identified and, if appropriate, a larger uncertainty may be assigned to these transitions.

4 Validation of Einstein Coefficients

Some Einstein coefficients entering the model are not available in the literature. An independent, experimentally based validation of the result of the structure calculations mentioned above is desirable and also needed before the data are included in databases like [10].

For the analysis of the EEDF, the Einstein coefficients are treated as nuisance parameters. Prior PDFs are assigned to them accounting for the uncertainty stated in [10] or specified according to a discussion of the BSRM results. The chosen uncertainty contributes to the uncertainty of the reconstructed EEDF and is accounted for by the probabilistic approach providing a non-Gaussian error propagation.

Figure 2 shows the result of the reconstructed EEDF from the spectroscopic data displayed in fig. 1, where the inferred EEDF is capable to describe all spectral features. The reconstructed EEDF agrees within the error margin with an EEDF derived from hybrid modelling of neon discharges [6, 7] up to energies of about 20 eV. The disagreement for higher energies was found to depend on the chosen parameterisation of the EEDF. This issue requires further studies, but the rate coefficients of the CRM that depend on the energy integral are barely affected by these deviations. Given this consistent result of the data reconstruction, i.e., within the relevant error margins, the emission spectrum is expected to contain also information on

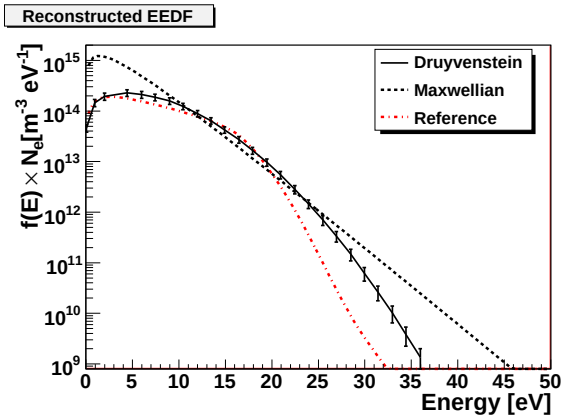

Fig. 2 Result for the reconstructed EEDF. On the ordinate axis the EEDF, multiplied with the reconstructed electron density, is plotted. The Maxwellian distribution shown for comparison is not able to reproduce the reference distribution from hybrid modelling [6].

the atomic data.

For the validation of the Einstein coefficients they are assigned a scale invariant prior probability distribution (Jeffrey's prior [11]). This means the results of the BSRM calculations are not included for these parameters and the width of the posterior distribution purely reflects the significance of the spectroscopic data for the considered coefficients.

The result of the analysis for the full set of available Einstein coefficients is shown in fig. 3 Displayed are the probabilities of the respective Einstein coefficients derived

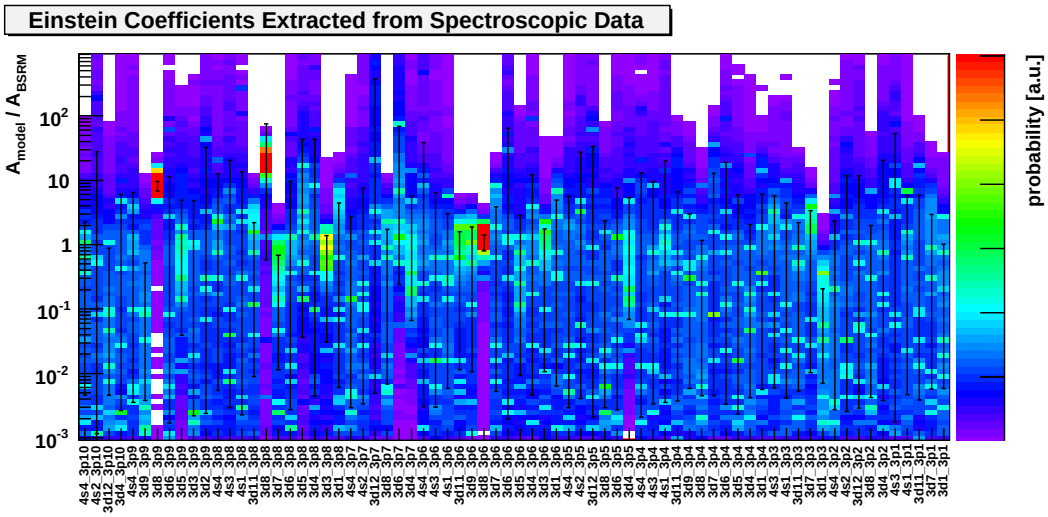

Fig. 3 The marginal distributions of various Einstein coefficients. The value of the coefficient is divided by the result of the BSRM calculations, with a value of 1.0 corresponding to full agreement (vertical left axis). The probability for a certain value of the coefficient to be true, given the measured data and model assumptions, is color-coded on the right, with violet/blue indicating a low and orange/red indicating a high probablity. The respective transition is given on the horizontal axis. The intervals depicted with the black lines show the root-mean-square variance of the distributions.

from the spectroscopic measurement and normalised to the BSRM results. There are a few lines that can be determined rather well, i.e., a narrow marginal posterior distribution is obtained. For example, a result consistent with the BSRM predictions is found for the $3d_8 \rightarrow 3p_6$ transition, whereas the value for $3d_8 \rightarrow 3p_8$ extracted from the spectroscopic data deviates significantly from the BSRM result. Further studies are needed to clarify whether this can be attributed to shortcomings in the spectroscopic analysis or problems with the BSRM calculations.

Figure 3 also reflects that the spectrum is not informative for many transitions, as indicated by a broad probability distribution for the respective transitions. Note that some transitions with a wavelength outside the measured spectrum are also included in the fit. They influence the spectrum through their effect on the collisional radiative model. However, as one might expect, these transitions have a particularly broad posterior distribution.

5 Conclusions

A full forward model for an emission spectroscopic measurement of a low-temperature plasma was used in a Bayesian data analysis. The electron energy distribution of the plasma was inferred and its error band resulting from uncertainties in the underlying atomic data was described. The extensive data set of excitation cross section obtained by recent close-coupling calculations allowed for the validation of Einstein coefficients for spontaneous emission, which have not been measured to date.

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