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Molecular Activated Recombination in Detached Recombining Plasmas

In a recent Letter, Ohno, Ezumi, Takamura, Krasheninnikov, and Pigarov (OETKP) presented an interesting study on the recombination of a dc plasma discharge in helium that is quenched by puffing hydrogen gas in the downstream region [1]. Their analysis is based on differences in upstream and downstream electron density $n_e$, temperature $T_e$, and He I line emission. They conclude that the puffing of hydrogen gas in the downstream region of the He plasma results in molecular recombination mechanisms becoming dominant. In agreement with observations on other experiments (e.g., Ref. [2]), OETKP find a transition from three-particle electron-ion recombination (EIR) to molecular activated recombination (MAR) when going from a pure He plasma to a He plasma with hydrogen gas added in the downstream region. Molecular activated recombination is a collection of molecular processes, of which $H_2(\nu) + e \rightarrow H^+ + H$, followed by $H^+ + H \rightarrow He + e$, and the formation of $HeH^+$ molecular ions followed by their dissociative recombination are mentioned by OETKP.

Our main objection concerns the line of evidence as presented in [1], especially as related to the determination of the downstream values of $T_e$. In the distinction between EIR and MAR processes, an accurate determination of $T_e$ is essential, as the rate of the EIR process has a $n_e^2T_e^{-9/2}$ dependence. The conclusions of OETKP regarding the dominance of MAR are based on a comparison of experimentally observed intensities of He I spectral lines with a collisional-radiative (CR) model [3]. The CR model is published as applied to a full hydrogen plasma but OETKP have apparently used it on He plasmas with hydrogen gas puffing. No reference or comments to this use of the model is supplied.

The used CR model is very sensitive to the assumed value of $T_e$. However, the authors give an estimate for the downstream value of $T_e$ (about 0.4 eV) only in the case of a pure He plasma, based mainly on an analysis of highly excited He state populations. This spectroscopic method may give a reasonable estimate for $n_e$ in the case of a purely noble gas plasma, but only if careful and absolute calibration of the spectroscopy setup and tomographic reconstruction of the line-averaged data are performed [4,5]. Estimation of $T_e$ is then possible under specific equilibrium conditions. OETKP fail to give any information regarding calibration and tomographic reconstruction and the figures show unprocessed spectral data: These are not presented in the usual way, i.e., as the natural logarithm of the state density per statistical weight vs the excitation energy of the radiating level. This makes an evaluation of the spectral data—and, thus, a judgement of the validity of the $T_e$ determination—by the reader impossible.

In the much more relevant situation of a He plasma with hydrogen gas puffing, however, no reasoning at all behind the adopted value of $T_e = 0.5$ eV is given. Actually, a $T_e$ determination by spectroscopic means may become impossible in this case, because (as OETKP indicate) the He I emission apparently disappears almost completely. To complicate things, the proposed molecular recombination mechanisms can strongly influence the excited state population [1,5] either directly or through influence on $n_e$ and $T_e$. However, OETKP apparently just assume that the $T_e$ value does not change when going from the pure He plasma to the hydrogen puffing case and use the same value for this crucial parameter in the model simulations for the two cases.

Alternative ways of determining $T_e$ are not mentioned in [1] and it remains obscure whether this is connected to the fact that, in another paper [6], anomalies with Langmuir probe $T_e$ measurements in the same experiment are reported.

In summary, OETKP reach their conclusion regarding the dominance of molecular processes in He plasmas with hydrogen gas puffing on the basis of unspecified estimates of the downstream $T_e$ (the crucial parameter in this case). The lack of absolute and local excited state population data (only a relative comparison to the model is made) adds to our doubts about the adopted method. In our view, an independent measurement of the electron temperature (e.g., by means of Thomson scattering) is essential to substantiate OETKP’s conclusions.

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