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Non-equilibrium nature of argon-based radiofrequency and microwave plasmas at atmospheric pressure evidenced by hyperfine optical emission spectroscopy

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In this work, the non-equilibrium nature of argon-based radiofrequency (RF) and microwave plasmas at atmospheric pressure is evidenced. In particular, rotational temperature (T_{rot}) and neural gas temperature (T_g) are found to be unequal in every condition tested, even though they are often assumed equal in such plasmas in the literature. Such a finding was made possible through the use of a hyperfine spectrometer developed and commercialized by LightMachinery Inc. Offering a 2 pm resolution over a simultaneous range of 25 nm, it was tuned to the 820-845 nm range in which the broadening of the Ar $2p_2 - 1s_2$ and Ar $2p_3 - 1s_2$ (Pashen notation) transitions is strongly affected by the neutral gas temperature. Therefore, for these emission lines, the experimental broadening was much smaller than the other broadening sources, ensuring a precise and accurate determination of T_g.

The microwave plasmas were produced inside a fused silica tube using a surfaguide-type wave launcher, while the RF plasmas were produced inside a fused silica tube placed between two electrodes. In both cases, an admixture of H_2O or N_2 were added to the argon flow in order to observe either the OH ($A^{2+} - X^2{}_i$) or the N_2^+ ($B2_u^+ - X2_g^+$) molecular systems. A rotational temperature was then calculated using the Boltzmann plot method. T_{rot} and T_g values were obtained every centimeter along the plasma columns. For the microwave plasma with an admixture of H_2O, T_g values of over 2000 K were obtained while T_{rot} values were in the 1400 K range. For the microwave plasma with an admixture of N_2, T_{rot} values were found to rise to ˜3200 K whereas T_g values only increased to ˜2400 K. The same discrepancies were found in the much colder RF plasmas (T_g˜400 K while T_{rot}˜515 K). Therefore, since the rotational temperatures did not equal the gas temperature in every condition tested, it is concluded that the rotational-translational equilibrium cannot not be assumed for RF and microwave argon-based plasmas at atmospheric pressure.

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