XXIX International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC2015) IOP Publishing
Journal of Physics: Conference Series **635** (2015) 032079 doi:10.1088/1742-6596/635/3/032079

A time-resolved spectroscopic study of UV/visible emissions from the He^*/He_2^* (n=3) manifold states: formation channels for fast He_2^* production

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We have investigated a fast formation channel yielding helium excited (bound) molecular states He_2^* from heavy-body conversion of n=3 manifold He^* atomic species, proposed by Frost *et-al* in 2001 (J.Phys.D, **34** 1569). Optical emission spectroscopy has been employed to measure the time-dependent intensity of ~20 atomic and molecular lines between λ=300-820nm from a short-pulse excited dielectric barrier discharge operating at 50-1000mbar. Correlations between atomic and molecular emission traces suggest that the singlet molecular state populations ($D^1\Sigma_u^+$, $E^1\Pi_g$, $F^1\Sigma_u^+$) can be attributed to conversion of the atomic $He^*(3^1S)$ state whilst the triplet molecular states are derived from the $He^*(3^3S)$ state.

Electronically excited (bound) He_2^* molecular states are known to play a major role in the kinetics of medium to high-pressure helium plasmas [1]. Key formation reactions include the three-body conversion of atomic $He^*(n=2)$ states and the di-electronic recombination of He_2^+ ions:

$$He^*(n=2) + 2He \Rightarrow He_2^*(^{1,3}\Sigma_{u,g}, ^{1,3}\Pi_{u,g}) + He \quad (1)$$
$$He_2^+ + 2e \Rightarrow He_2^* + e \quad (2)$$

In 2001, Frost *et-al* [2] reported transient (τ<200ns) broad-band (molecular) emissions in the visible following pulsed laser state-selective pumping into the higher $He^*(n=3)^{1,3}P$ levels. They proposed the existence of a previously neglected fast formation channel of molecular states involving the creation of a short-lived (τ<5ns) intermediate activated He_2^* complex (denoted by $He_2^{*‡}$) via two-body collisions (3). $He_2^{*‡}$ subsequently undergoes fast radiative (4) and/or collisional (5) stabilization:

$$He^*(n=3, ^{1,3}P) + He \Rightarrow He_2^{*‡} \quad (3)$$
$$He_2^{*‡} \Rightarrow He_2 + h\nu \quad (4)$$
$$He_2^{*‡} + He \Rightarrow He_2^* + He \quad (5)$$
$$He_2^* \Rightarrow He_2 + h\nu \quad (6)$$

They also noted that the reactions (4)-(5) would compete with other known relaxation processes of $He_2^{*‡}$ including associative ionisation and predissociation. To our knowledge, no follow-up studies of these observations have been reported, and the specific molecular states in (4) and (5) have yet to be identified. Moreover, due to weak LIF signals and low spectral resolution (Δλ~4nm), Frost *et-al* were unable to resolve the PQR branches within their emission bands to allow individual lines to be matched to known He_2^* transitions.

We have employed a short-pulse excited dielectric barrier discharge (DBD) to generate transient (~200ns) pulsed plasmas in helium at pressures of 50-1000mb. The experimental system and techniques are described in detail in [3]. DBD excitation is well suited to studies of heavy-body collisions in high pressure plasmas due to the intrinsic self-quenching of the discharge current pulse which limits the plasma electron density and subsequent electronic collisional processes e.g.(2) which may compete with heavy-body driven reactions.

The time-resolved intensities of ~20 selected He atomic lines and molecular bands between λ=300-820nm, originating from the n=3,4 manifolds, have been recorded using an Acton VM504 monochromator (Δλ~0.1nm resolution) and SRS430 multichannel analyzer (Δt~5ns resolution) with the detection system operating in photon counting mode. The principal results of our study show that band emission originating from both singlet and triplet He_2^* n=3 manifold molecular states ($D^1\Sigma_u^+$-$B^1\Pi_g$, $E^1\Pi_g$-$A^1\Sigma_u^+$, $F^1\Sigma_u^+$-$B^1\Pi_g$, $d^3\Sigma_u^+$-$b^3\Pi_g$, $e^3\Pi_g$-$a^3\Sigma_u^+$, $f^3\Sigma_u^+$-$b^3\Pi_g$) occurs concurrently with the atomic line emission from $He^*(n=3)$ states during the plasma excitation phase (when He_2^* formation via He_2^+/e recombination can be neglected). This suggests that a fast formation channel of molecular species is operative. Using a kinetic model based on conversion via an intermediate $He_2^{*‡}$ state, the time-dependent emission curves from atomic/molecular transitions have been further analysed to determine possible correlations. Our results suggest that population transfer to the singlet molecular states ($D^1\Sigma_u^+$, $E^1\Pi_g$, $F^1\Sigma_u^+$) can be attributed to heavy-body conversion of the atomic $He^*(3^1S)$ state, thus linking $He_2^{*‡}$ to high-v excited states of $F^1\Sigma_u^+(3d\sigma,3s)$ and $^1\Sigma_g^+(4f\sigma,3s)$. Similarly, the triplet molecular states are populated via $He^*(3^3S)$ conversion.

References

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