

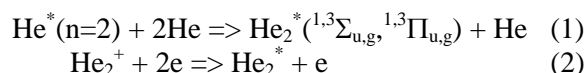
A time-resolved spectroscopic study of UV/visible emissions from the He^{*}/He₂^{*} (n=3) manifold states: formation channels for fast He₂^{*} production

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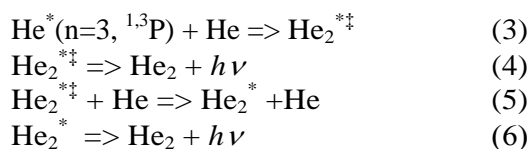
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We have investigated a fast formation channel yielding helium excited (bound) molecular states He₂^{*} from heavy-body conversion of n=3 manifold He^{*} atomic species, proposed by Frost *et-al* in 2001 (J.Phys.D, **34** 1569). Optical emission spectroscopy 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 (D¹Σ_u⁺, E¹Π_g, F¹Σ_u⁺) can be attributed to conversion of the atomic He^{*}(3¹S) state whilst the triplet molecular states are derived from the He^{*}(3³S) state.

Electronically excited (bound) He₂^{*} 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 dielectronic recombination of He₂⁺ ions:



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 He₂^{*} complex (denoted by He₂^{*†}) via two-body collisions (3). He₂^{*†} subsequently undergoes fast radiative (4) and/or collisional (5) stabilization:



They also noted that the reactions (4)-(5) would compete with other known relaxation processes of He₂^{*†} 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 He₂^{*} 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 He₂^{*} n=3 manifold molecular states (D¹Σ_u⁺-B¹Π_g, E¹Π_g-A¹Σ_u⁺, F¹Σ_u⁺-B¹Π_g, d³Σ_u⁺-b³Π_g, e³Π_g-a³Σ_u⁺, f³Σ_u⁺-b³Π_g) occurs concurrently with the atomic line emission from He^{*}(n=3) states during the plasma excitation phase (when He₂^{*} formation via He₂⁺/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 He₂^{*†} 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 (D¹Σ_u⁺, E¹Π_g, F¹Σ_u⁺) can be attributed to heavy-body conversion of the atomic He^{*}(3¹S) state, thus linking He₂^{*†} to high-v excited states of F¹Σ_u⁺(3dσ,3s) and ¹Σ_g⁺(4fσ,3s). Similarly, the triplet molecular states are populated via He^{*}(3³S) conversion.

References

- [1] F. Emmert *et-al* 1988 *J.Phys.D.* **21** 667
- [2] M.J. Frost *et-al*, 2001 *J.Phys.B* **34** 1569
- [3] R.J.Carman *et-al*, 2010 *J.Phys.D* **43** 025205

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