CARS Measurements of Vibrational Distribution Function in Nanosecond Pulsed Plasmas

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Picosecond Coherent Anti-Stokes Raman Scattering (CARS) has been used for studies of vibrational energy dynamics in a diffuse single filament pin-to-pin discharge in pure N_2 and air. The discharge produces a filament with approximate dimensions 2-3 mm (diameter) x 10 mm (height) using a pair of 7.5 mm bare copper spherical electrodes.

Results for N_2 at P = 100 Torr are summarized in Fig. 1, which shows experimental and Master equation modeling predictions for total vibrational quanta/molecule in levels v = 0 – 8 for which population was observed, as well as discharge total specific energy loading, and modelling predictions for energy stored in vibration. It can be seen that experimentally approximately 30% of coupled pulse energy is loaded into the N_2 vibrational mode which is in qualitative agreement with the modelling prediction of approximately 45%. However it can also be seen that the total vibrational quanta/molecule rises rapidly after the termination of the discharge pulse, by a factor of between ~two and three for time delays after discharge initiation in the range ~10–200 microseconds. This suggests that additional energy, up to a maximum of ~70% of the coupled discharge pulse energy, is loaded into the vibrational mode after the discharge pulse. This trend is clearly at variance with the kinetic model, which predicts the number of quanta to remain nearly steady for up to t~200 μsec after the pulse, since V-V exchange among nitrogen molecules conserves the total number of quanta in the N_2 vibrational mode. At longer time delays slow relaxation is observed due to a combination of V-T relaxation, mass diffusion, and convection.

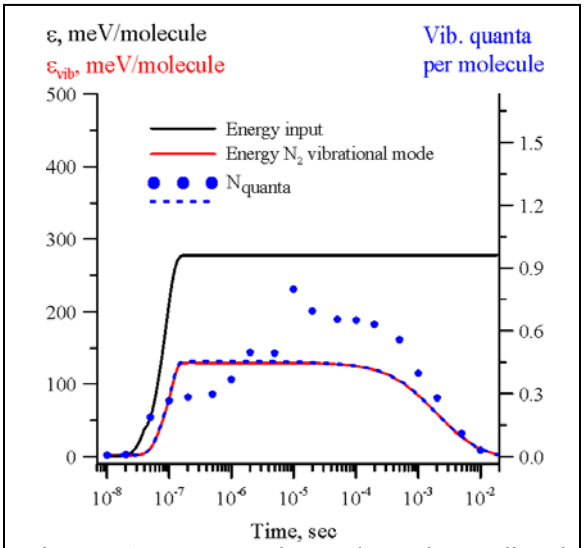

Figure 1 – Experimental and predicted temporal evolution of vibrational energy and total vibrational quanta in 100 Torr N_2 nsec pulsed discharge.

The results summarized in Fig.1 , as well rotational/translational temperatura and more detailed measurements of the temporal evolution of the nitrogen vibrational distribution function, suggests that a significant fraction of energy initially stored in excited electronic species and dissociation products by electron impact is subsequently transferred to the vibrational mode of the ground $N_2(X^1\Sigma,v)$, rather than to heat, during collisional processes such as the energy pooling process, $N_2(A)+N_2(A)\rightarrow N_2(B,C)+N_2(X,v)$. Similar results are obtained in air at P = 100 Torr and from spontaneous Raman scattering measurements in which detectable population was observed in vibrational levels as high as v = 12.

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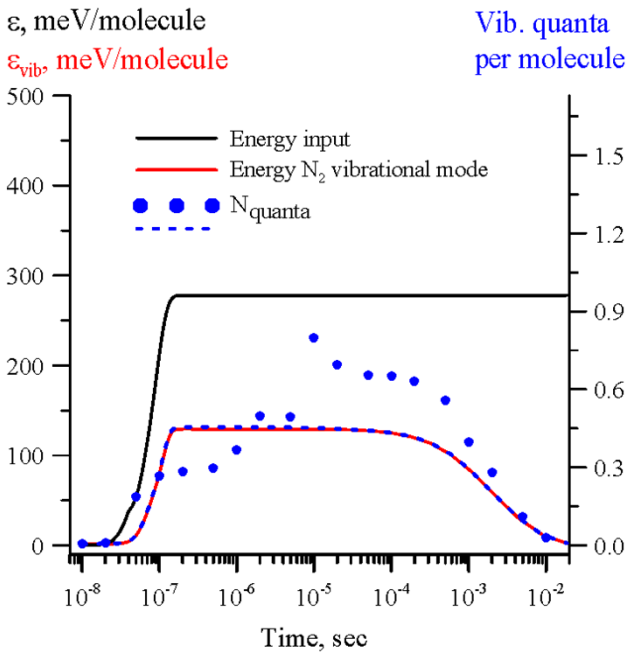
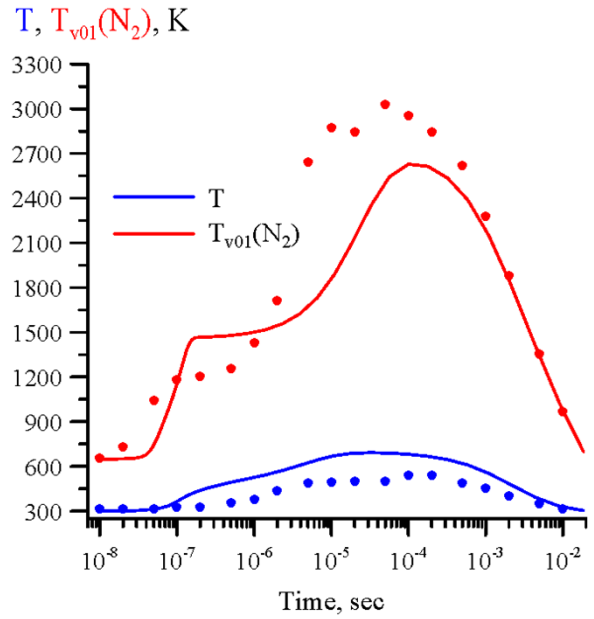
ENERGY LOADING AND DECAY OF N_2 VIBRATION IN PULSED N_2 AND AIR PLASMAS: NEW PHYSICS?

- CARS (Coherent Anti-Stokes Raman Scattering) laser diagnostics demonstrated that ~40% of coupled energy from ~100 ns discharge pulse is initially loaded into N_2 vibrations (v=0-8 detected) by electron impact.
- N_2 vibrational energy increases to ~80% of coupled energy for time scales ~10-200 μsec after discharge pulse (symbols). Model (solid lines) predicts vib energy to be constant, under-predicts 1st level vib temperature, and over-predicts rotational temperature.
- Mechanism for $N_2(X,v)$ population after the pulse is unknown – quenching of $N_2(A,B,C,a)$ quenching or N atom recombination are possibilities.



- Vibrational Energy of N_2 (v=0-8)



- 1st Level Vibrational and Rotational Temperature

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Particle Charging in Argon-Hydrogen Plasmas

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Capacitively coupled plasma (CCP) RF discharges are used for gas phase synthesis of silicon nanocrystals. Typically, SiH_4 diluted in ≈95% argon is flowed through the discharge chamber at a pressure of a few Torr [1]. The plasma dissociates SiH_4, creating radicals that react and form uniformly sized, crystallized silicon nanoparticles which are collected downstream. Interaction of the plasma with the nanoparticles plays a role in determining their surface and bulk properties. The particles acquire a steady state negative charge and are impacted by energetic ions in the plasma. Processes such as rearrangement of atoms in the particle to form a crystal lattice and termination of the atoms on the surface depend on the impaction energies of the ions.

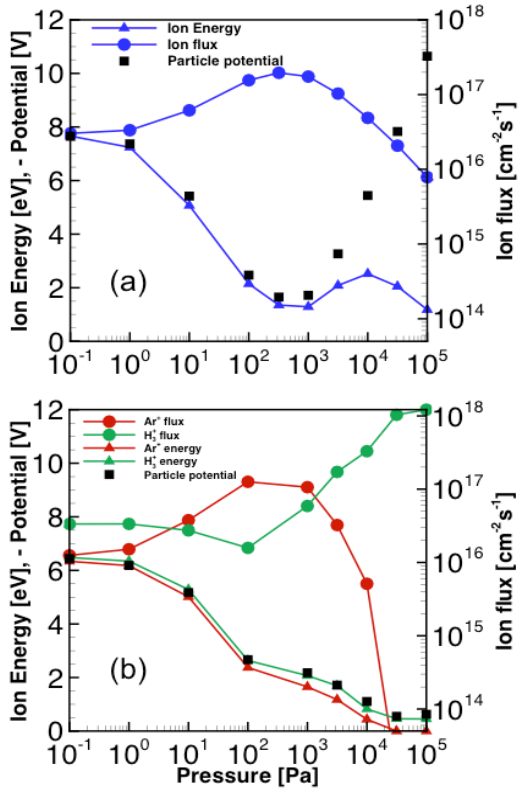

Figure 1: Particle potential, ion flux, and mean ion impact energy for a 500 nm diameter nanoparticle in plasmas containing (a) $Ar^+ = 1\times10^{10}$ cm^{-3} and (b) $Ar^+ = 5\times10^{9}$ cm^{-3}, $H_3^+ = 5\times10^{9}$ cm^{-3}.

Trace amounts of H_2 (≈2%) are produced in the discharge due to dissociation of SiH_4 and subsequent gas phase reactions. The H_2 is ionized and the plasma may consist of comparable fractions of heavy Ar ions and light hydrogen ions [2]. Plasma-particle interactions in the presence of comparable fractions of argon and hydrogen ions were investigated.

An analytical model was developed to calculate particle charging in plasmas and the results of the model agree with Monte Carlo simulations. The steady state particle potential, ion flux and average ion impact energy for a 500 nm particle are shown in Fig. 1. The discharge consists of either only Ar^+ ions or equal densities of Ar^+ and H_3^+ ions. In the pure argon discharge the particle potential reduces (becomes less negative) at intermediate pressures due to enhancement of the ion current, and increases (more negative) at higher pressures as the ion current is inhibited by collisions. In the presence of both Ar^+ and H_3^+ ions, the ion flux of each species to the nanoparticle peaks at different pressures due to the difference in their ion-gas collision characteristics. The H_3^+ ion current peaks at a higher pressure compared to the Ar^+ current. The presence of H_3^+ ions causes a reduction in the particle potential (less negative) over a wide pressure range, and so a reduction in the average ion impact energy.

References

[1] L. Mangolini, Nano Letters **5**, 4 (2005).
[2] A. Bogaerts, and R. Gijbels, Spectrochemica Acta Part B: Atomic Spectroscopy **57**, 6 (2002).

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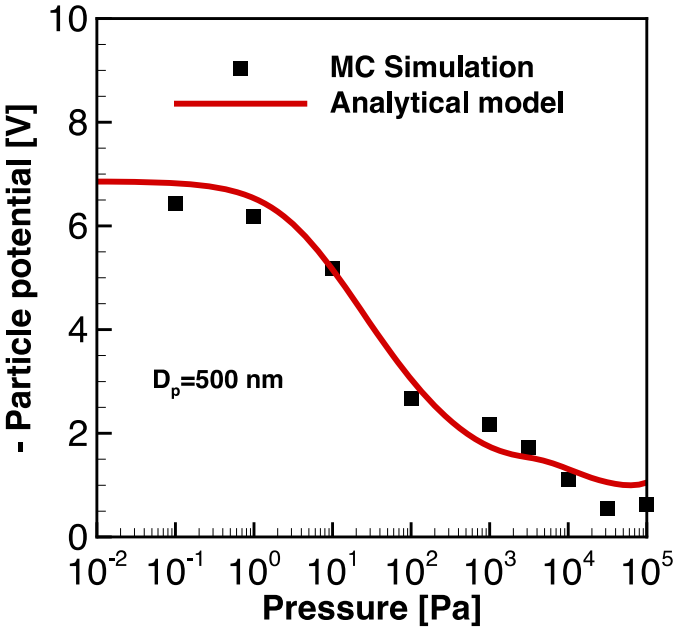
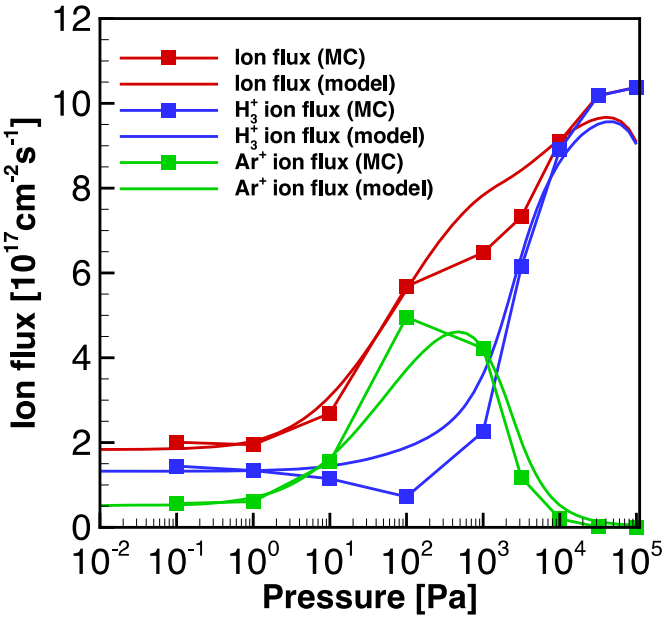
PARTICLE CHARGING IN Ar-H_2 PLASMAS

- An analytical model calculates steady state nanoparticle charge in a plasma containing multiple positive ions and compares well with Monte Carlo simulations.
- In Ar-H_2 discharges containing equal number of Ar^+ and H_3^+ (5×10^9 cm^{-3}) ions, flux of each species to the particle depends on the individual ion-gas collision characteristics and so fractional current is pressure dependent.



- Steady state particle potential



- Total and individual ion fluxes to particle