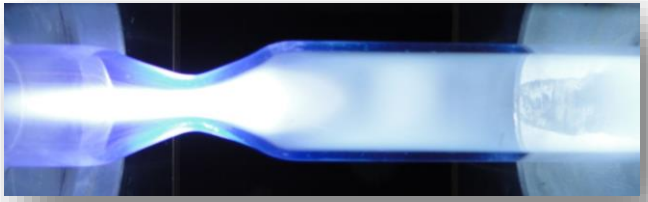
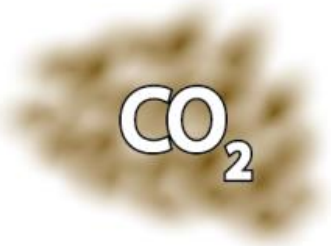
Tailoring plasma vibrational excitation to enhance the energy efficiency of chemical conversions

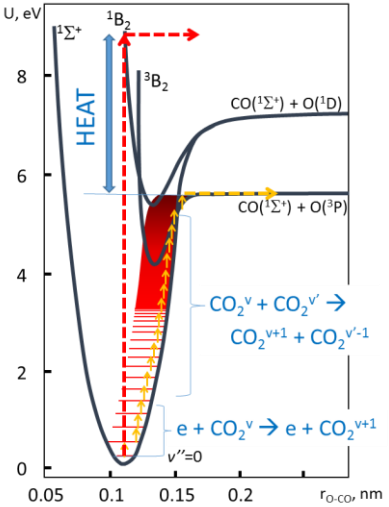
Gerard van Rooij, Waldo Bongers, Richard van de Sanden

G.J.VANROOIJ@DIFFER.NL

DIFFER, Dutch Institute For Fundamental Energy Research



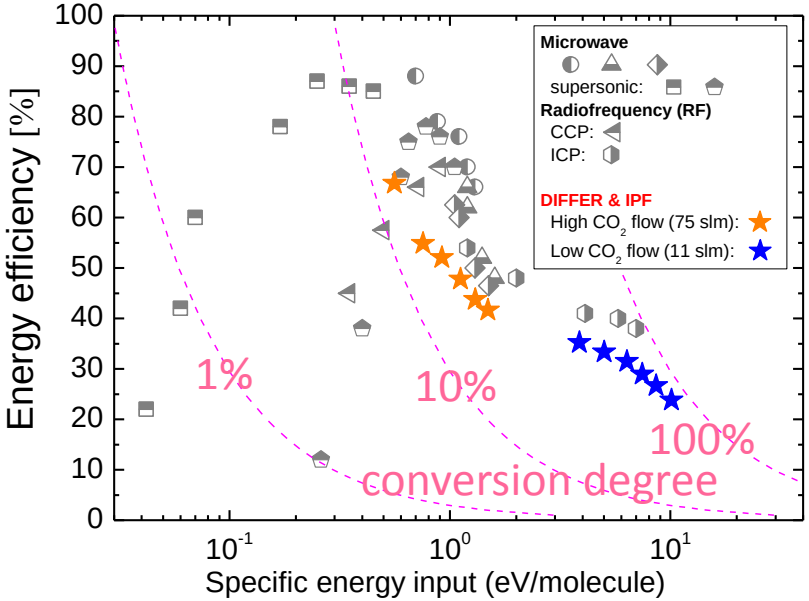


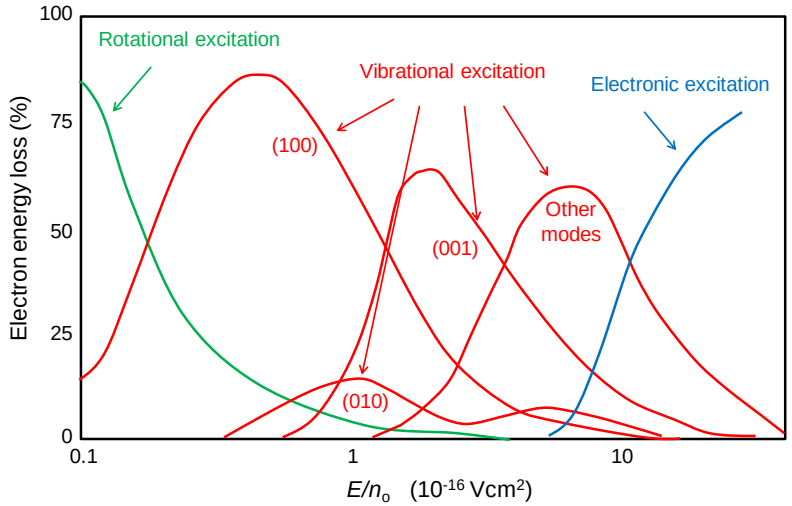


The challenge

Can the plasma phase be used to effectively beat thermodynamic equilibrium conversions?

The frontier

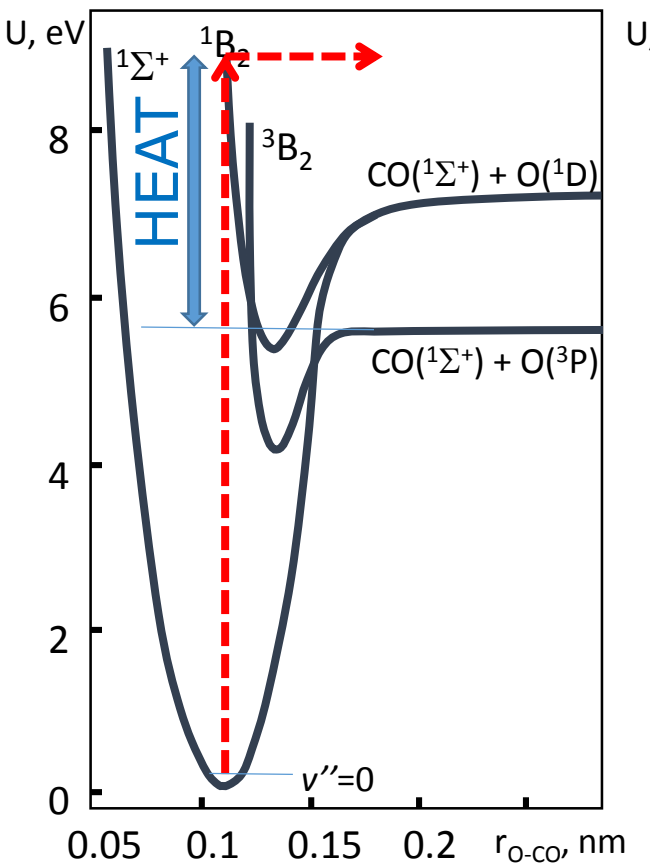




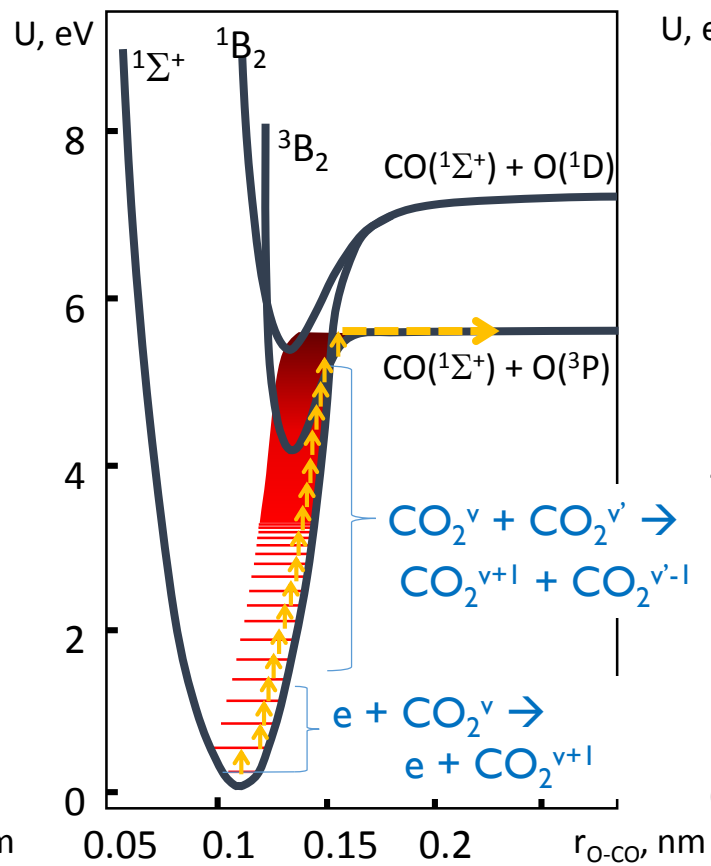
- CO_2 dissociation efficiencies of up to 90% inspiration for employing plasma phase for chemical conversions
- The mechanism: channeling the energy to the bond to be broken (rather than to translation or ionization) → *non-equilibrium*
- The challenge: demonstration of economical viability
- sustainable energy sources important driver: *electricity* & *intermittency*

Understanding - relevant molecular physics for CO_2

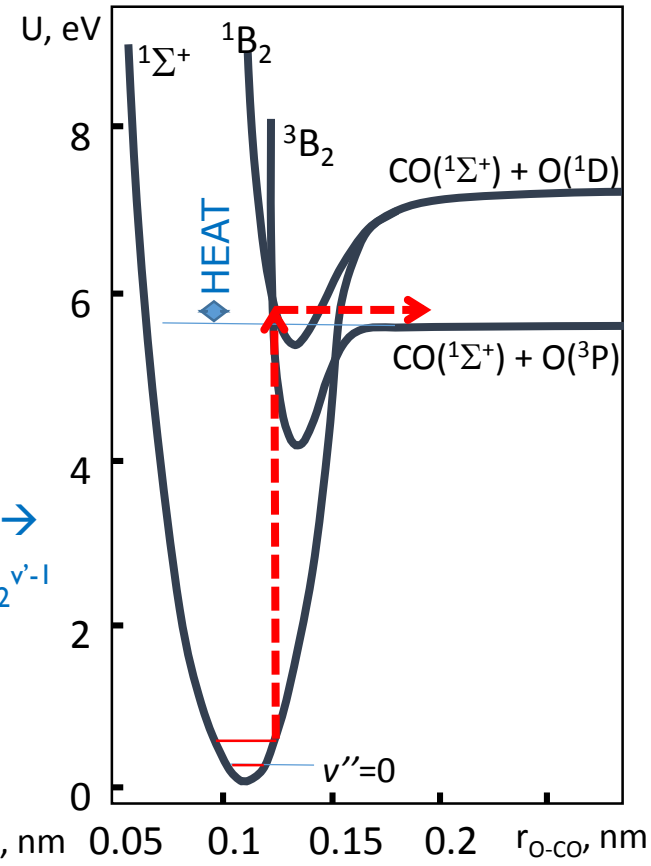
Dissociative excitation



Vibrational ladder climbing

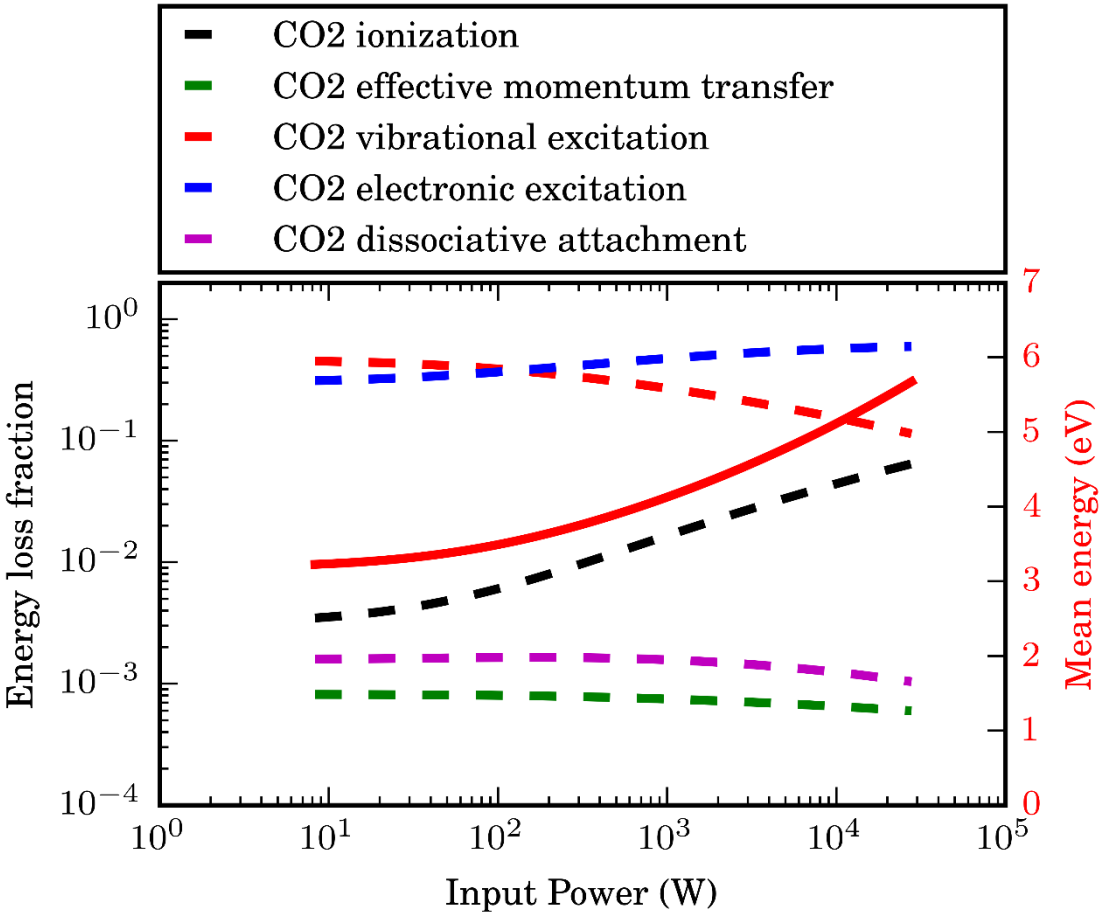


Vibrationally enhanced dissociative excitation



Van Rooij et al., Faraday Discussions, 2015, DOI: 10.1039/C5FD00045A

Caveat: MW-plasma not inherently promoting v-excitation

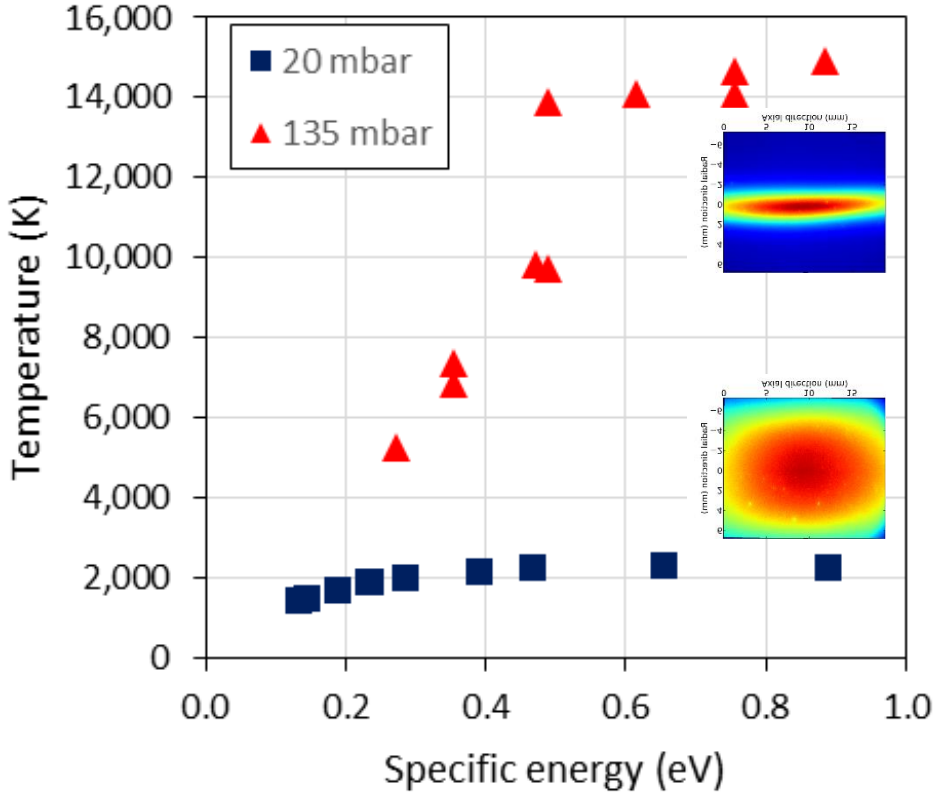


BOLSIG analysis

Dissociative recombination lowers ionization life time and promotes electronic excitation

Van Rooij et al., Faraday Discussions, 2015, DOI: 10.1039/C5FD00045A

Experiments confirm high gas temperatures



Rayleigh measurements

Diffuse discharge mode yields lower temperatures, contracted mode better efficiency

Van Rooij et al., Faraday Discussions, 2015, DOI: 10.1039/C5FD00045A

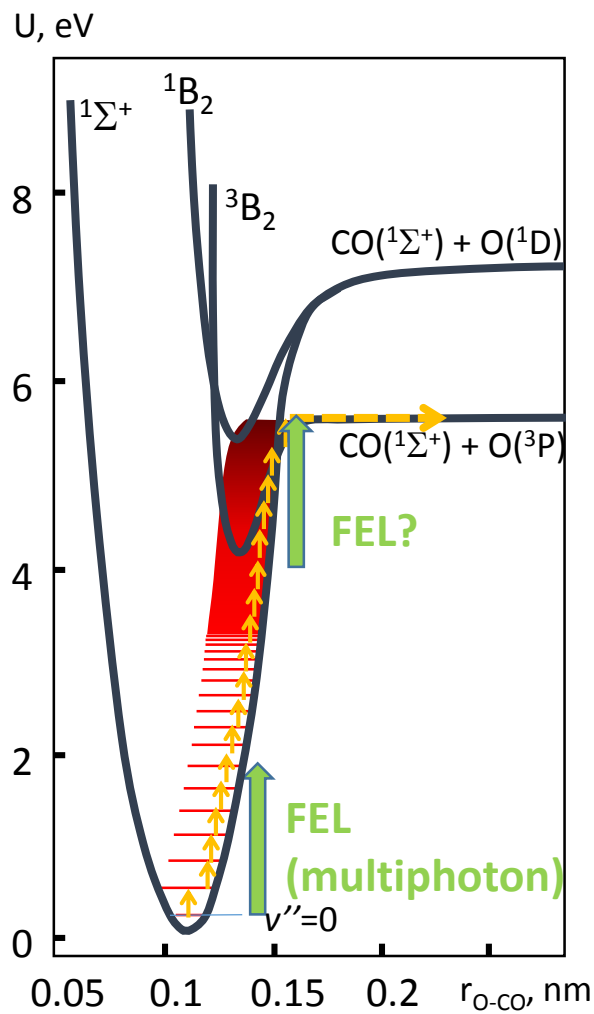
Advancing the frontiers

1. Improve understanding of vibrational exciation → free electron laser experiments

2. Tailor EEDF by additives (low ionization potential alkali metals)

3. Optimize vibrational enhanced dissociative excitation: reactor geometry / pulsed operation

1. Improving understanding with free electron laser

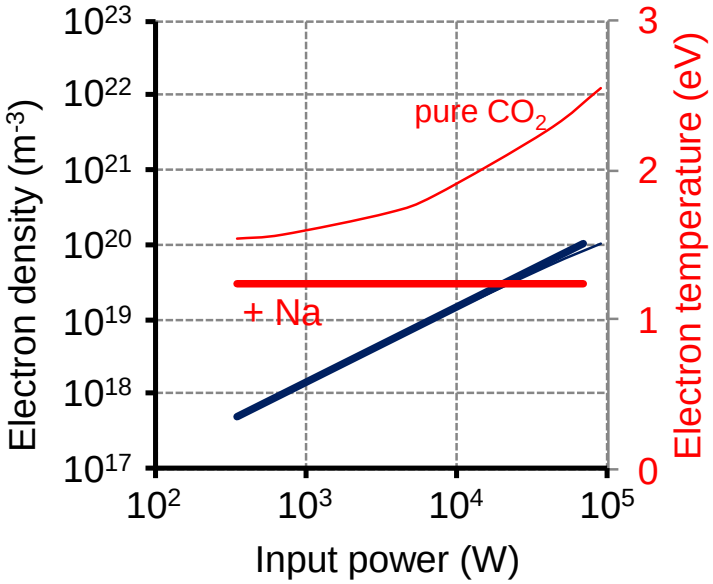


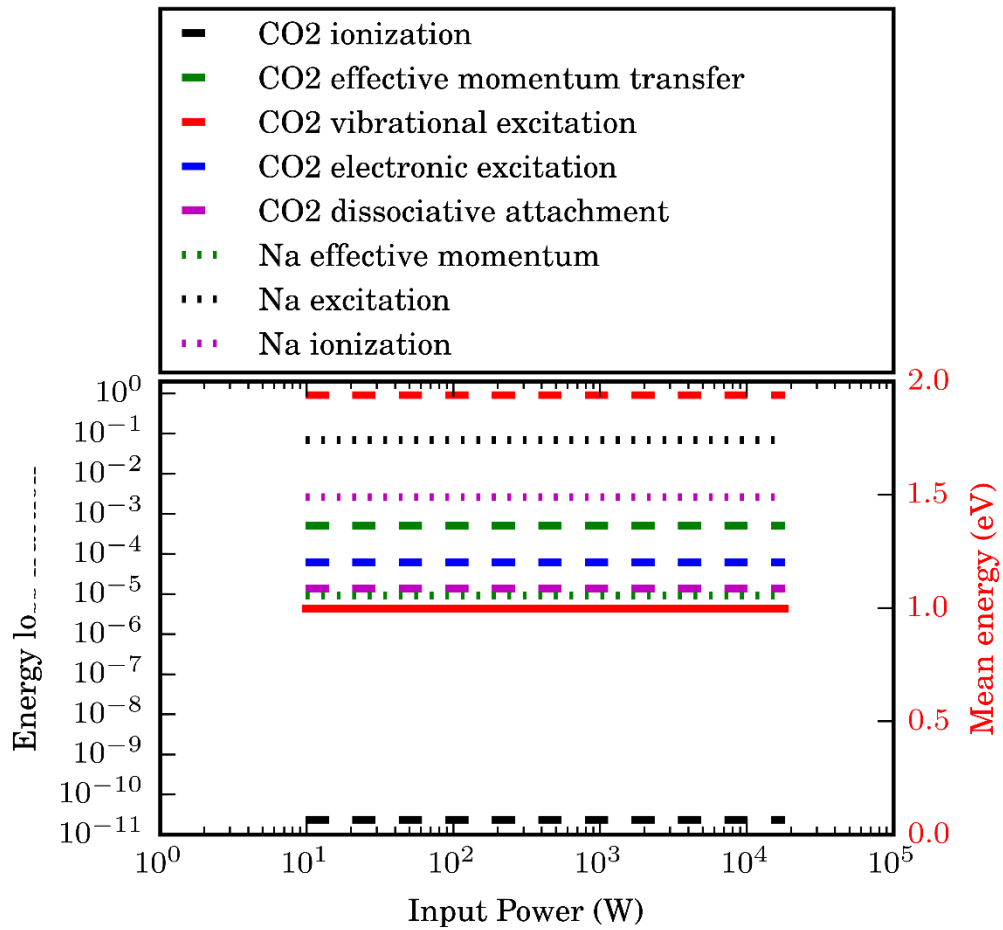
Perturb intrinsic vibrational distribution to investigate:

- dynamics of *v*-perturbation
- effect on CO formation & dynamics of *v*-*v* relaxation
- role of different vibration modes

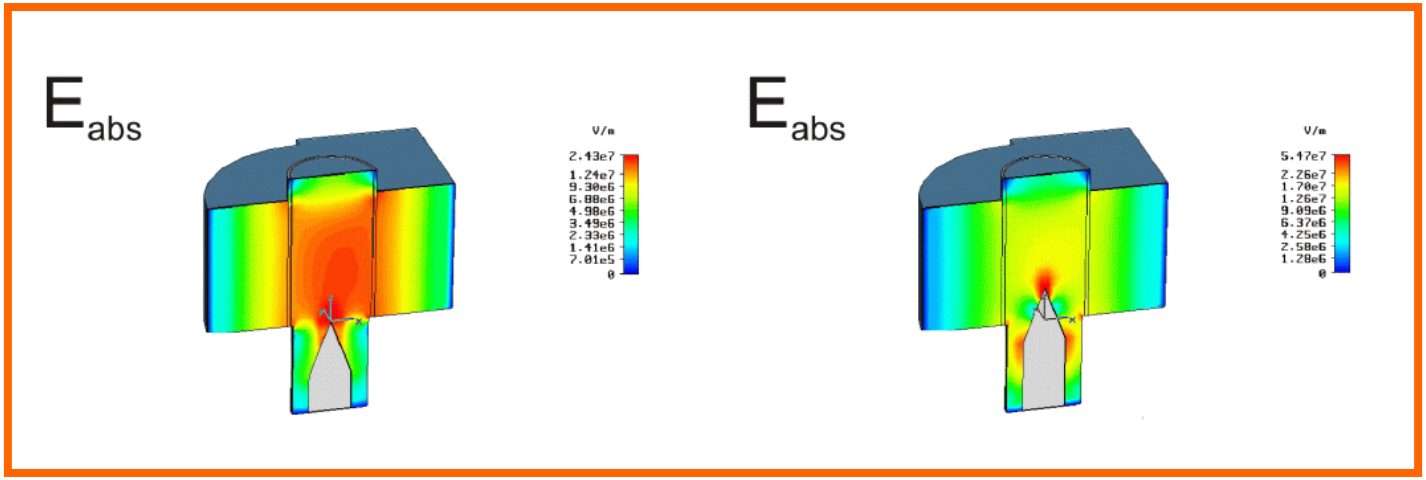
What happens with energy deposited into the vibrational manifold?

2. Tailoring EEDF with low ionization potential additives





3. Vibrational enhanced dissociative excitation



Shape EEDF through spatio- & temporal electric field control (emphasizing electronic excitation)

Impact

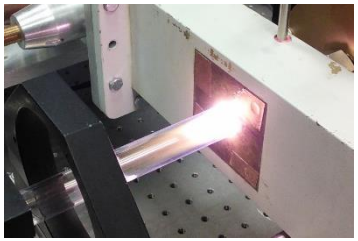
- Sustainable energy flexibility (intermittency)

- CO_2 to synthetic fuel (sector integration)

- Nitrogen fixation (fertilizers)

- Carbon coupling in methane (chemicals)

Acknowledgement



Dirk van de Bekerom, Niek den Harder, Teofil Minea, Jose Palomares, Adelbert Goede, Michael Gleeson, Stefan Welzel

DIFFER
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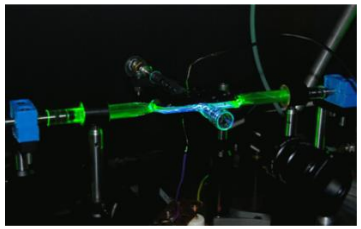
Giel Berden, Lex vd Meer, Britta Redlich

Radboud University Nijmegen



Richard Engeln, Bart Klarenaar

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Projects (NWO, STW-Alliander, TKI) running in collaboration with:

Shell

HOST BIO-ENERGY INSTALLATIONS

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