Data needed for modeling Low Temperature Plasmas (LTPs)

Leanne Pitchford
Laboratory des Plasma et Conversion d'Energie (LAPLACE)
CNRS & University of Toulouse III, France

pitchford@laplace.univ-tlse.fr

Outline

This talk will focus low-temperature, non-equilibrium, collision dominated plasmas created in electric discharges operating at pd (pressure x dimensions) such that volume, not surface, processes dominate.

1) Context
2) Modeling LTPs - identification of data needs
3) Presentation of LXCat
4) Other data needs for modeling & actions
5) Conclusions

Technologies based on LTPs – some examples

Electrical energy

PLASMA

Radiant energy
Source of photons

- Lamps
  - Lighting
  - Water purification
  - Sterlization
- Visualisation
  - Plasma display panels
- Lasers

Kinetic energy
Source of charged particles

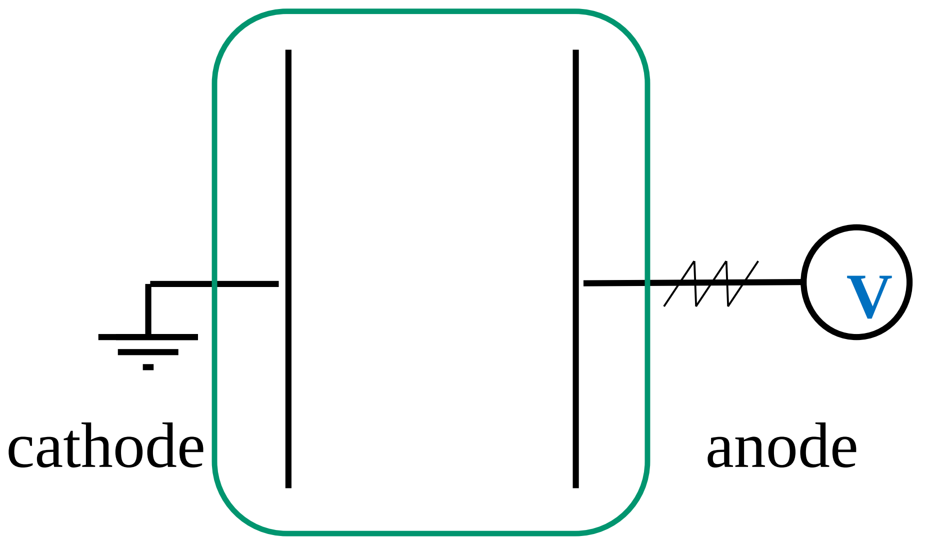
- Ion sources
  - Ion implantation
  - Propulsion
  - Ion sources for ITER
  - Elemental analysis of solids
- Electron beams
  - X-ray sources
- Switches, current interrupters

Chemical energy
Source of reactive species

- Surface processing
  - Microelectronics
  - Functionalisation of surfaces
- Volume processes
  - Pollution control
  - Chemical conversion
- Biological applications

Generation of LTPs

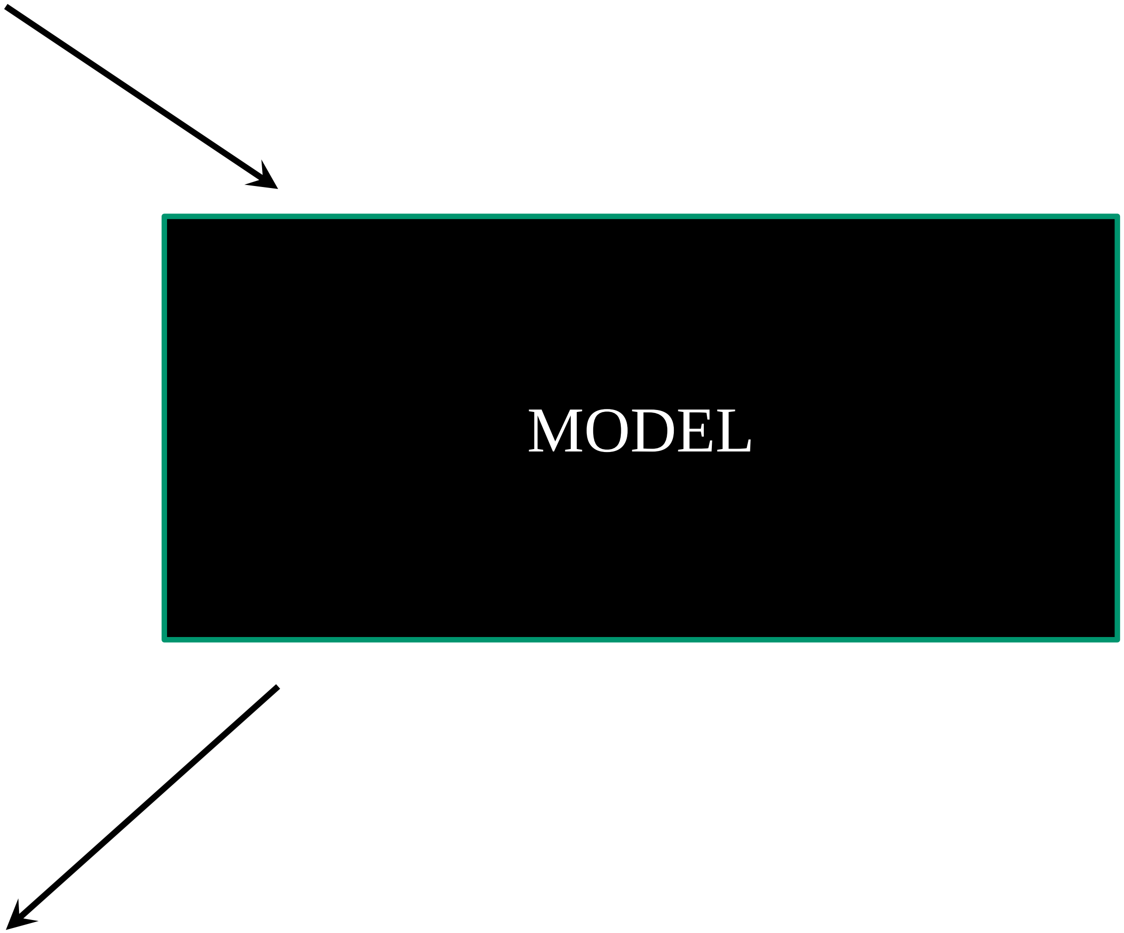
LTPs can be generated most simply by applying a voltage (> V_b) beween two electrodes, separated by a gas gap.



For a range of conditions: **"T_e" >> T_i = T_g** => the electrons are the vector through which electrical energy is deposited in the gas through collisions, leading to excitation, dissociation, & ionization.

Overview of discharge models

INPUT: gas composition and pressure; geometry; circuit,…



OUTPUT: **E**, n_e, n_+, T_g, neutral species densities,… as functions of **x**,t.

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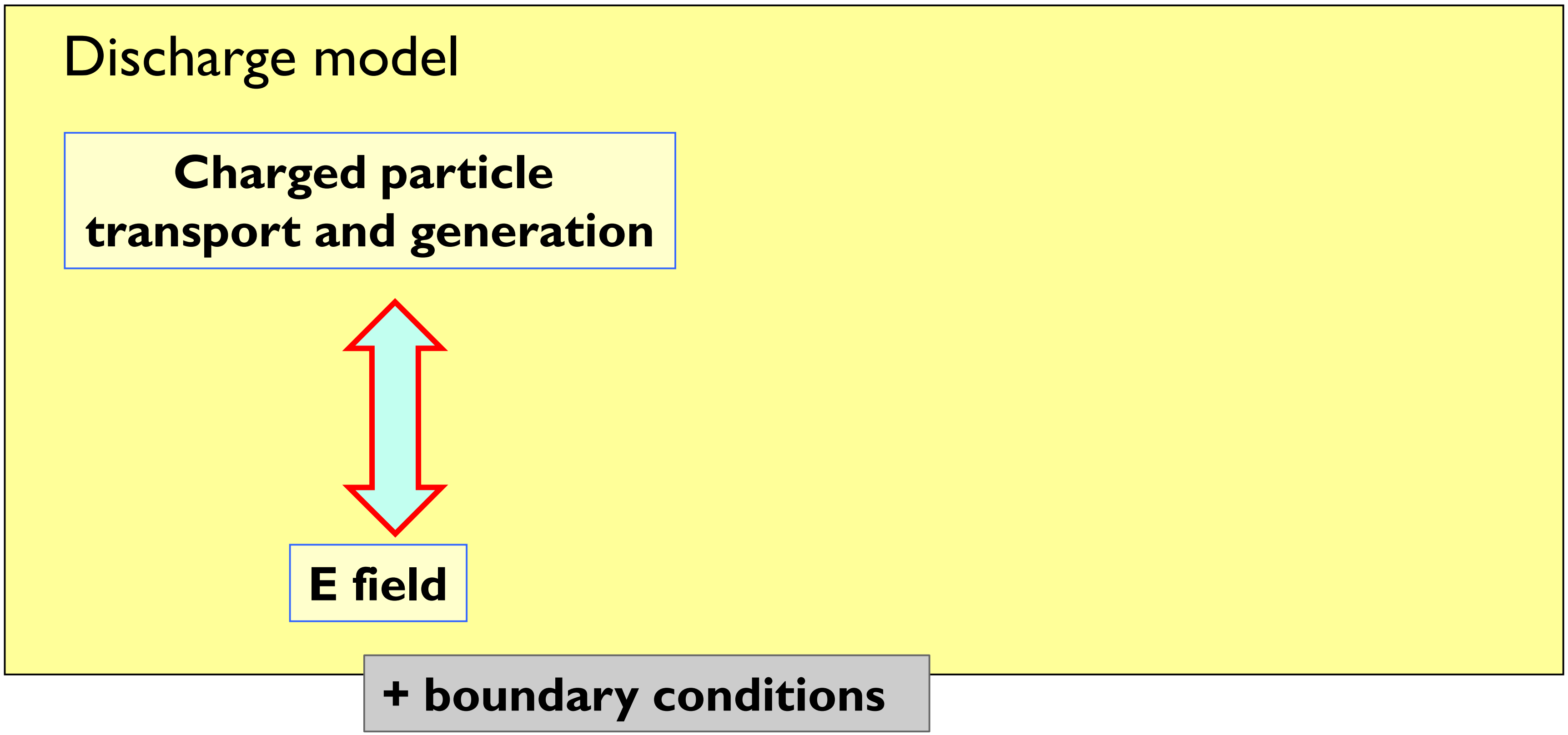

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Overview of discharge models

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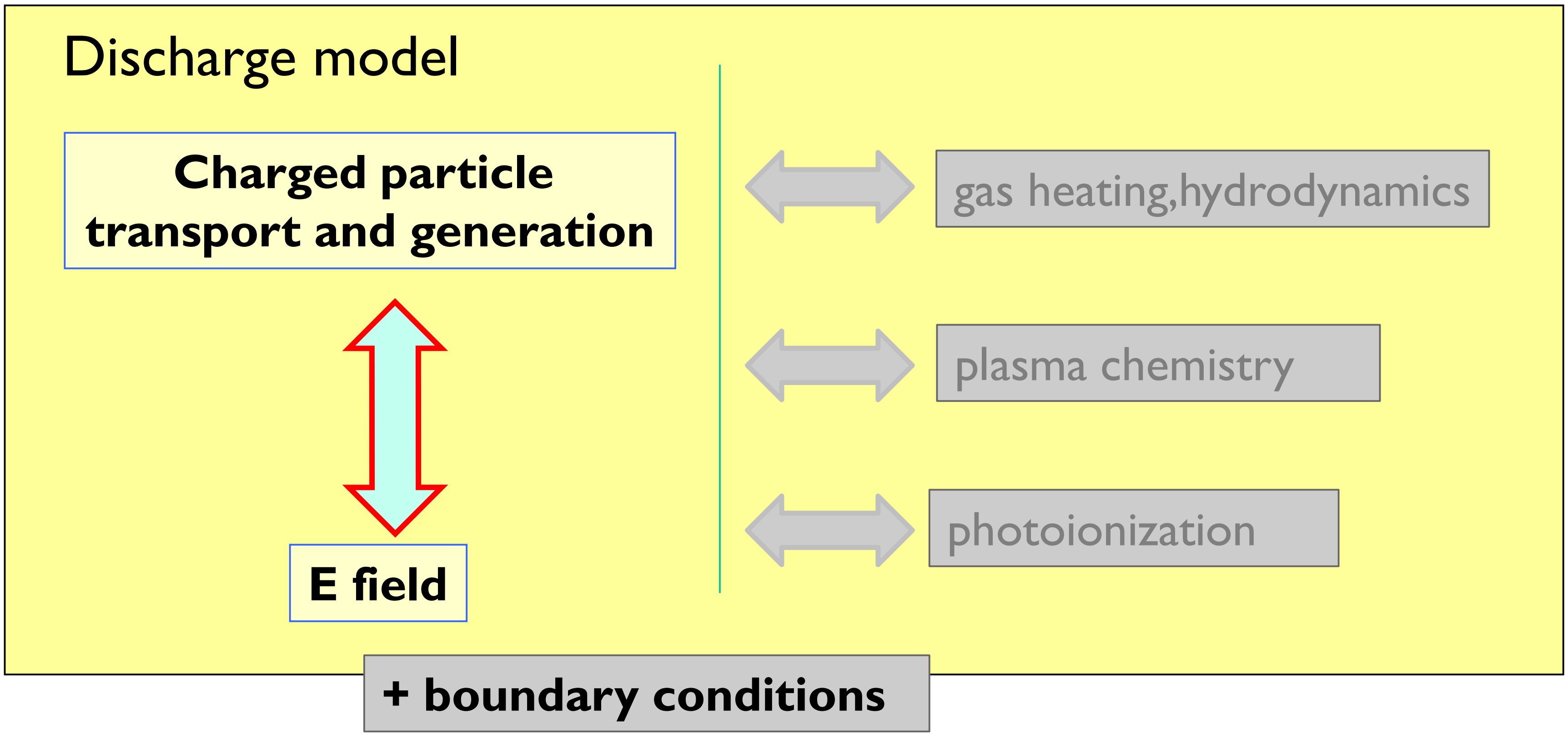


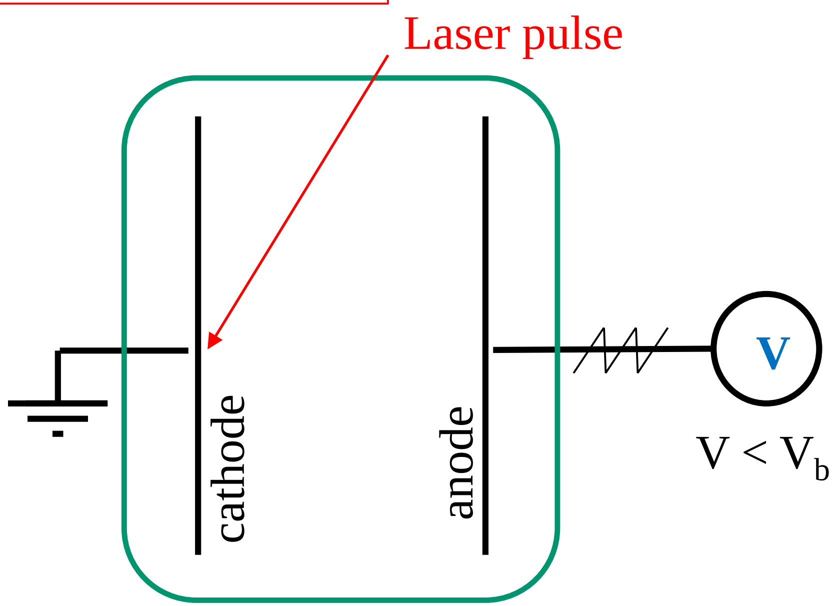
OUTPUT: **E**, n_e, n_+, T_g, neutral species densities,… as functions of **x**,t.

Modeling charged particle transport & generation

LEVEL OF DESCRIPTION	DATA NEEDS
PARTICLE MODELS (Boltzmann or Monte Carlo simulations)	**Electron- and ion-neutral scattering cross sections** "Complete" sets of electron/ion - neutral cross sections
HYBRID MODELS	Certain aspects of ion or electron behavior are treated with particle models; other aspects with fluid models.
FLUID MODELS (multi-fluid)	**Electron and ion transport coefficents** – mobility and diffusion coefficients - **and ionization rates.**

Measurements of transport coefficients (one example)

Experimental set-up



- $\mathbf{E} = E_z$ = constant, except near radial boundaries.
- Neutral gas number density, N.
- Electrons, generated at t=0, z=0, are accelerated to the anode ($a = -eE_z/m$), undergoing collisions with neutrals along the way.
- E/N is a good parameter.
- "Swarm"

Results

Analysis of current waveforms at the anode yields drift velocity, longitudinal diffusion coef, and ionization rate coef.

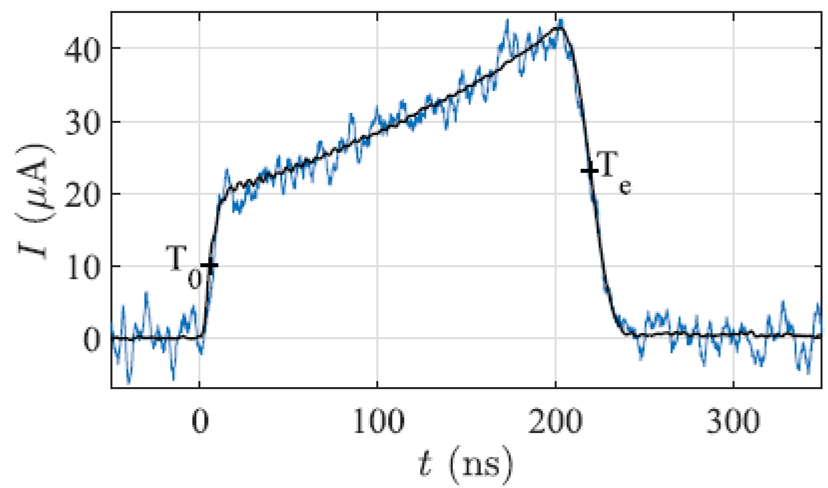


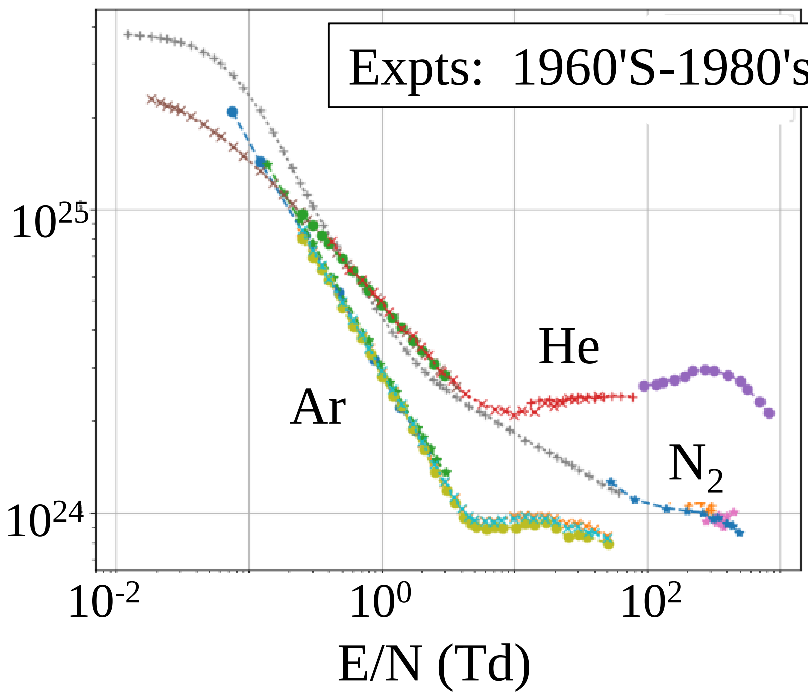
FIG. 4. Measurement with a typical best SNR in N_2 at $p = 10$ kPa, $d = 25$ mm, and $(E/N) = 110$ Td. (blue solid line) Single waveform and (black solid line) average of 200 single waveforms. (+) T_0 and T_e of average.

Haefliger & Franck, Rev. Sci. Instrum. 2018

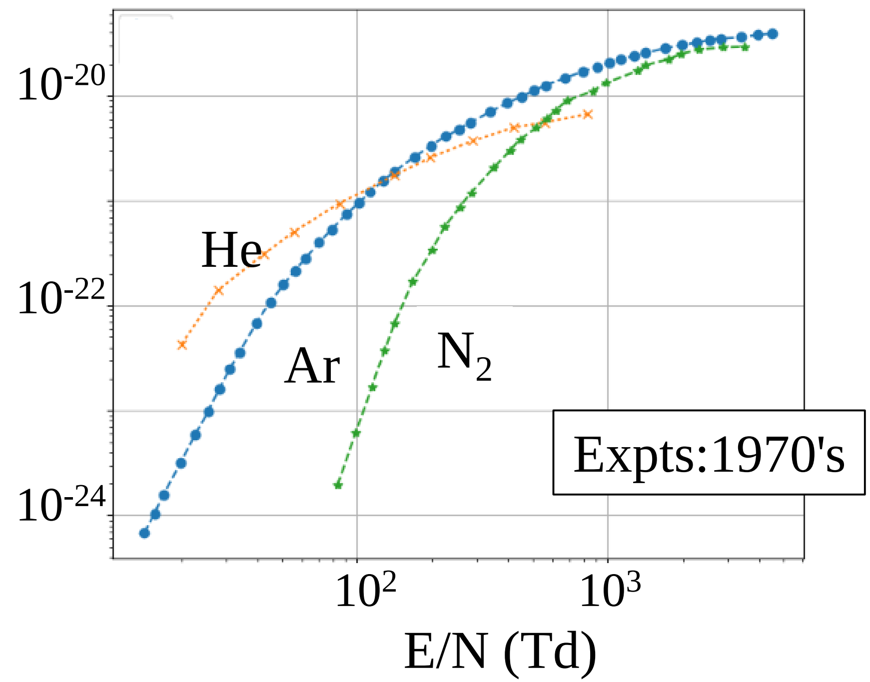
Examples of measured electron transport coefficients

See Dutton database on www.lxcat.net for data with references

Reduced mobility (μN) in /m/V/s)

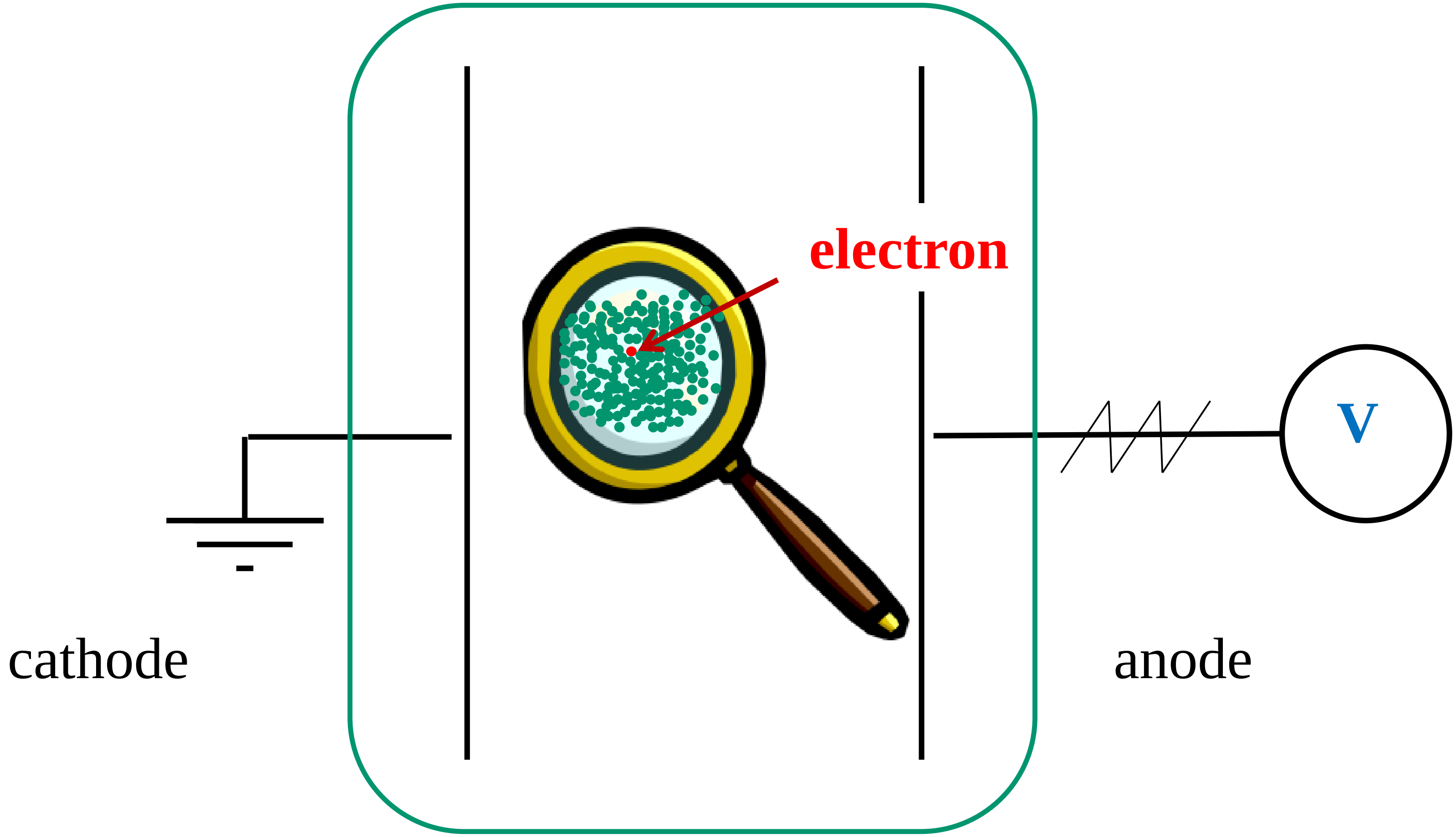


Townsend ioniz. coef (α/N) in m^2)



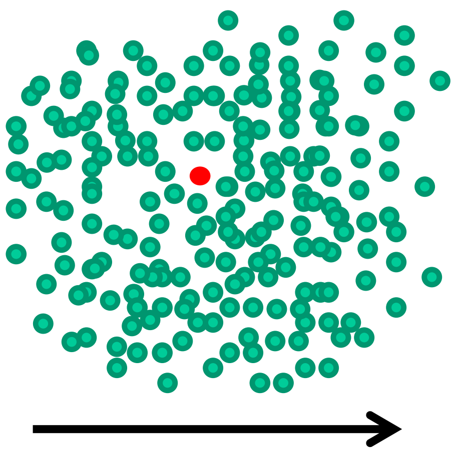
- The SI Unit for E/N is Townsend (Td), 10^{-21} V m^2.
- The relation between drift velocity and reduced mobility : $v_d = \mu N \times E/N$.
- The relation ionization rate coefficient and Townsend coef.: $\alpha/N = k_i/v_d$.
- In Ar at 1 Td, the thermal energy is ~ 2.4 eV, whereas the directed energy is ~ 2.10^{-2} eV.
- Other measureable transport data include $D_T N$, $D_L N$, D_T/μ.

LTPs – a closer look



Boltzmann transport equation

f(r,v,t) = electron velocity probability distribution function



a = -e**E**/m

$$\frac{\partial f}{\partial t} + \vec{v} \cdot \nabla f + \vec{a} \cdot \nabla f = NC\left[f \right]$$

transport in r

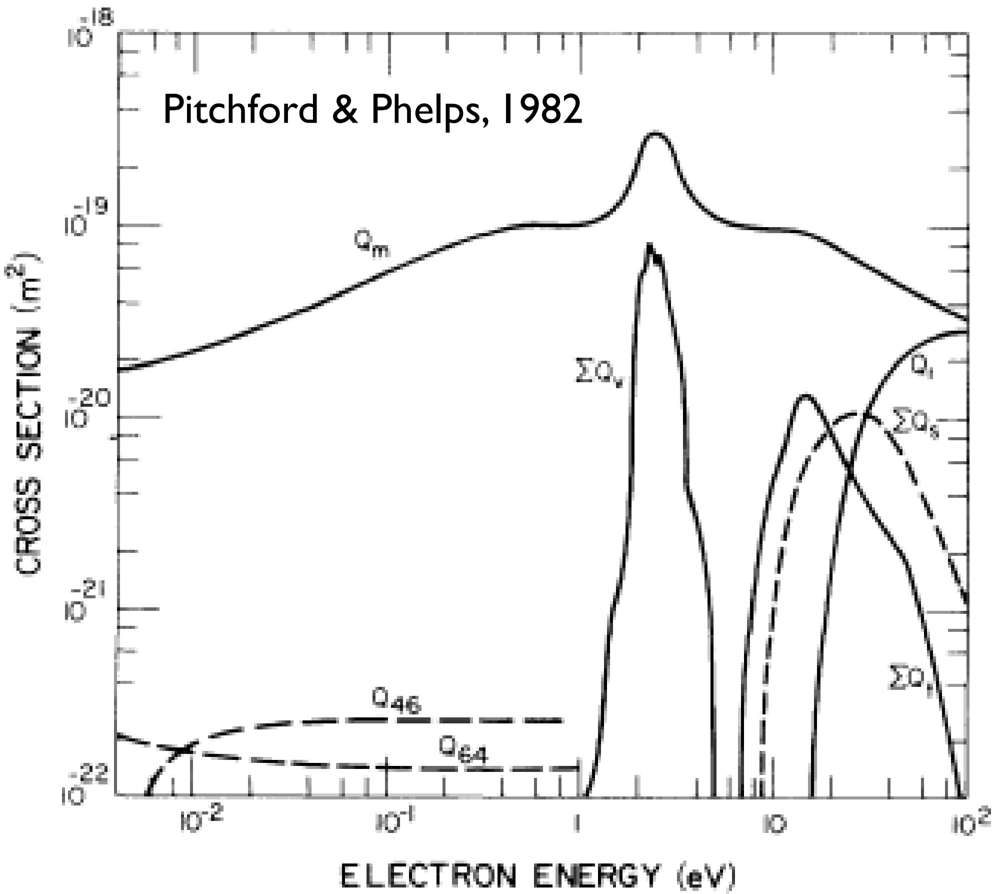
transport in v

Collisions

where $\int f(\boldsymbol{r}, \boldsymbol{v}, t) d\boldsymbol{v} = n_e(\boldsymbol{r}, t)$

Electron transport & rate coefficients are integrals over *f*(**r**,**v**,*t*).

Input data required for Boltzmann equation solutions



Types of collisions:

- *Elastic*

Recoil energy loss, momentum transfer ($Q_{m,el}$)

- *Inelastic*

Discrete energy losses due to excitation of rotational, vibrational and electronic states ($Q_{k,T}$)

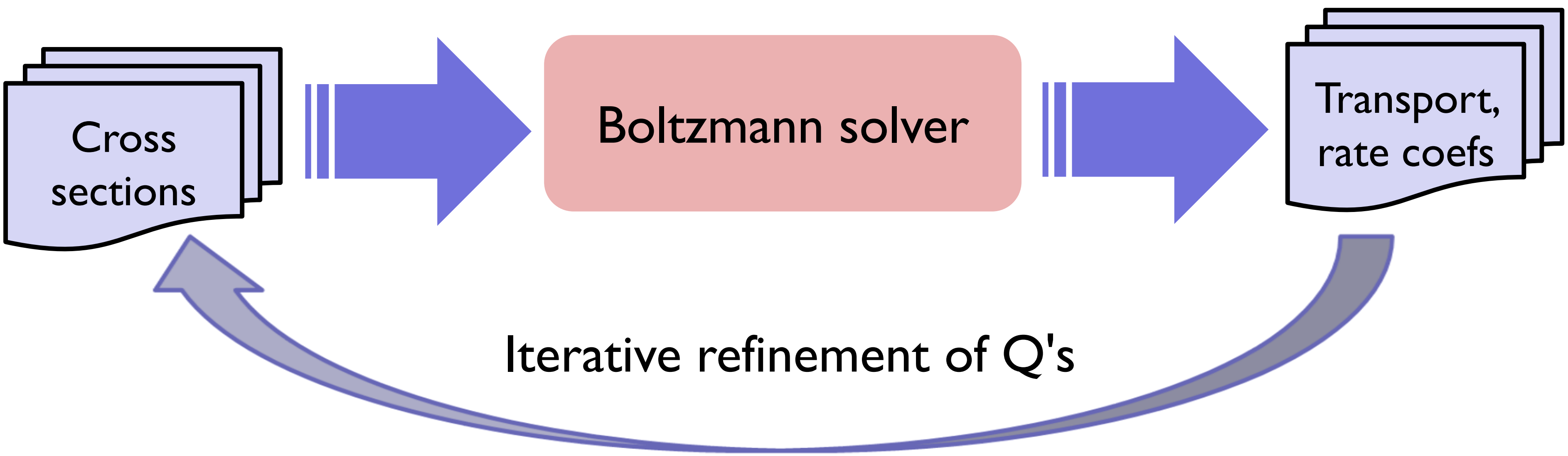
- *Ionization*

Two electrons exit the collision event ($Q_{i,T}$, energy sharing)

Solutions of the Boltzmann equation

Analytical solutions exist only for simplified cases, elastic scattering only, power law $Q_{m,el}$ (eg Maxwellian, Druyvesteyn).

An important research activity has been the development of ***numerical solution techniques*** which enable direct connection of fundamental AMO data & plasma transport.



The LXCat project (initiated at LAPLACE in 2010)

LXCat is a web-based, community-wide project for the curation of

data needed in modeling low-temperature plasmas

Electron + neutral
cross sections / oscillator strengths/ swarm parameters

Ion + neutral
cross sections / interaction potentials / swarm parameters

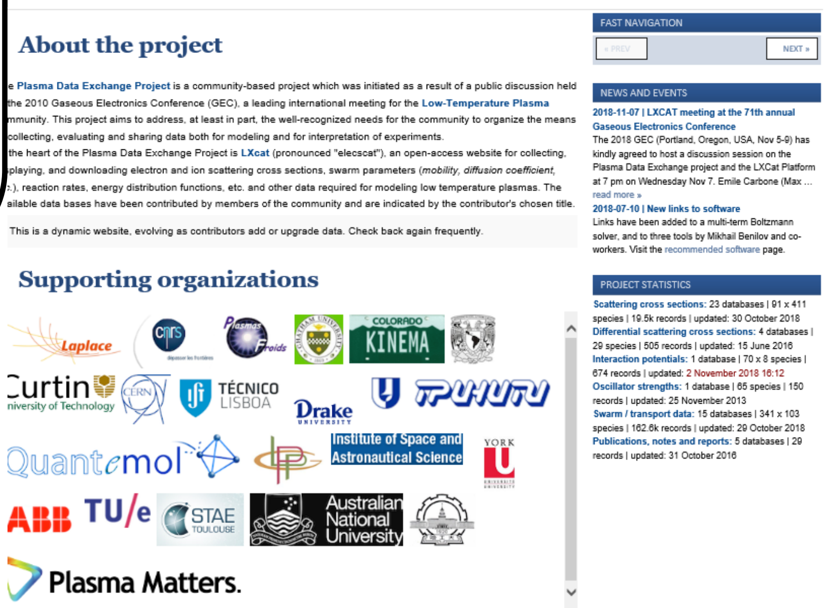
Plasma-surface interactions

Plasma chemistry

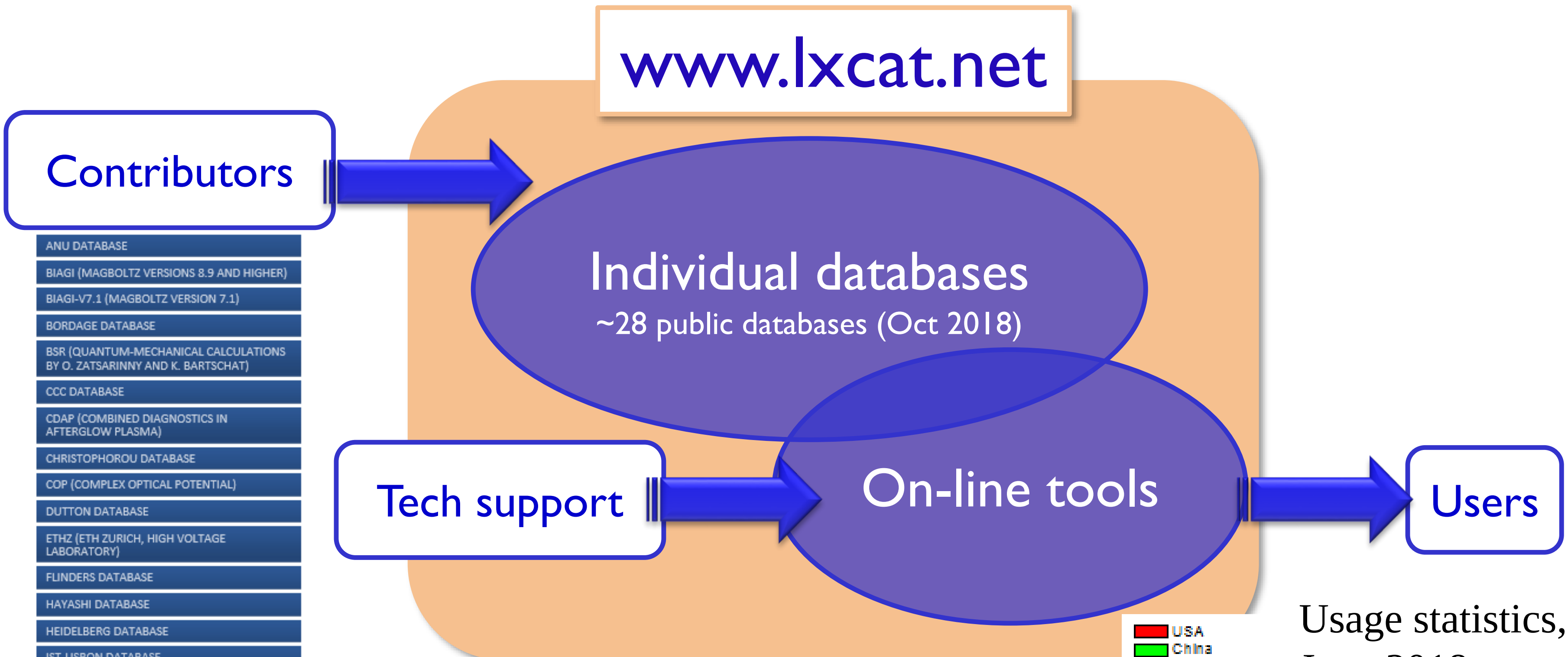
Radiation

LXCat

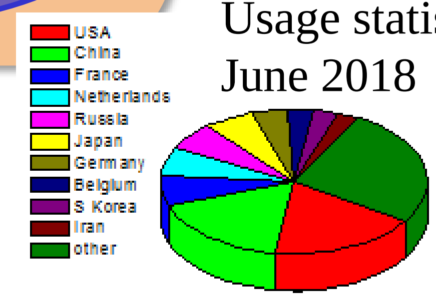
ELECtron (and ion) SCATtering



LXCat structure – databases & on-line tools



- ANU DATABASE
- BIAGI (MAGBOLTZ VERSIONS 8.9 AND HIGHER)
- BIAGI-V7.1 (MAGBOLTZ VERSION 7.1)
- BORDAGE DATABASE
- BSR (QUANTUM-MECHANICAL CALCULATIONS BY O. ZATSARINNY AND K. BARTSCHAT)
- CCC DATABASE
- CDAP (COMBINED DIAGNOSTICS IN AFTERGLOW PLASMA)
- CHRISTOPHOROU DATABASE
- COP (COMPLEX OPTICAL POTENTIAL)
- DUTTON DATABASE
- ETHZ (ETH ZURICH, HIGH VOLTAGE LABORATORY)
- FLINDERS DATABASE
- HAYASHI DATABASE
- HEIDELBERG DATABASE
- IST-LISBON DATABASE
- ITIKAWA DATABASE
- LAPLACE (MEASUREMENTS AFTER 1975)
- MORGAN (KINEMA RESEARCH & SOFTWARE)
- NGFSRDW DATABASE
- PHELPS DATABASE
- PUECH DATABASE
- QUANTEMOL DATABASE
- SIGLO DATABASE
- TRINITI DATABASE
- UBC DATABASE
- UNAM DATABASE
- UT (UNIVERSITY OF TARTU)
- VIEHLAND DATABASE



Usage statistics, June 2018

S Pancheshnyi *et al.*, *Chem. Phys.* **398**, 148 (2012)
LC Pitchford et al, *Plasma Process. Polym.* **14,** 1600098 (2017)

Contributors to the LXCat project (to Sept 2018)

Website conception: S Pancheshnyi, (France /Switzerland)

Scattering cross sections (compilations, quantum calculations, measurements) : MC Bordage, V. Puech, LC Pitchford (France); SF Biagi, D Brown, J Tennyson (UK); K Bartschat, WL Morgan, AV Phelps, J. Stephens, L Viehland, MC Zammit, O Zatsarinny (USA); LL Alves, C Ferreira, V Guerra (Portugal); NA Dyatko, IV Kochetov, AP Napartovich (Russia); Y Itikawa (Japan); I Bray, S Buckman, M Brunger, L Campbell, D Fursa, McEachran (Australia); A Stauffer (Canada); RK Gangwar, L Sharma, R Srivastava (India)

Oscillator strengths: C Brion (Canada)

Transport/rate coefficients (compilations, measurements) : L Viehland, AV Phelps (USA); S Chowdhury (France), J de Urquijo (Mexico); LL Alves, V Guerra (Portugal); Christophorou (Greece); A Chachereau, CM Franck, P Haefliger, A Hoesl, M M Hildebrandt (Germany); L Rabie (Switzerland); X-M Zhu (China); I Jogi (Estonia)

Ion-neutral interaction potentials: L Viehland (USA)

Initial website development: S Pancheshnyi, (France /Switzerland); B Chaudhury (India),

Tech support: W Graef, D Mihailova, J van Dijk (The Netherlands); M Hopkins, B Yee (USA), Pancheshnyi

Outreach: K Bartschat (USA), E Carbone (Germany), LC Pitchford (France), Y-K Pu (China)

On-line Bolsig+ : GJM Hagelaar (France); S Pancheshnyi (Switzerland)

Servers: Eindhoven Technical Univ. & Univ Toulouse

Complete sets of e/neutral cross sections available on LXCat

….for electron scattering in COLD gases, $Q_{k,T}$ for inelastics, $Q_{m,el}$ for elastic

Atomic gases

Ar, C, Cu, H, He, Hg, Kr, Mg, N, Na, O, Xe

Diatomic gases

CH, CO, Cl_2,D_2, F_2, H_2, HCl, N_2, NO, O_2

Polyatomic gases

C_2H_2, C_2H_4, C_2H_6, C_3H_6, C_3H_8, CCl_2F_2, CCl_4, CF_4, CH_4, CH_3, CH_4, CHF_3, CO_2, H_20, N_2O, SF_6, SO_2, Si_2H_6, $Si(CH_3)_4$, SiH_4

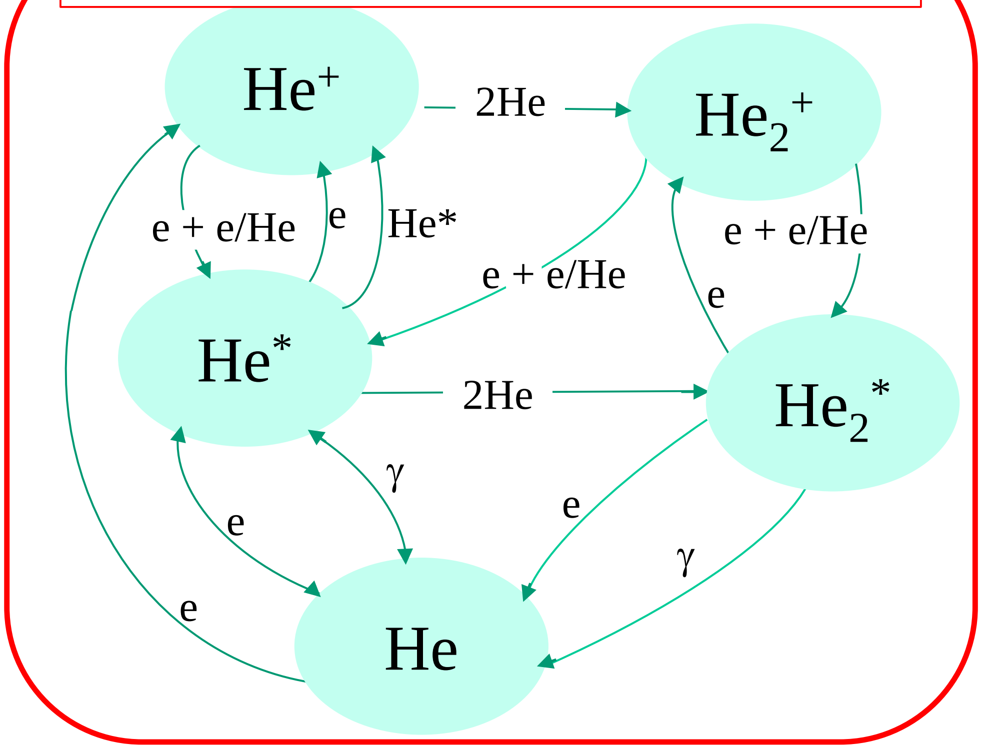
ATTENTION: LXCat does not recommend data.

On-line : **BOLSIG+ Lite** => Boltzmann solver for quick on-line calculations of transport and rate coefficients in gas mixtures, comparisons with measurements

Other data needs include plasma chemistry

The consensus in the Low Temperature Plasma Community is that there is a need to define a strategy to develop, validate (to the extent possible) and distribute ***reaction mechanisms*** for some common gas mixtures, analogous to the effort already made in the combustion community (GRE-Mech).

Example: a basic mechanism for helium, suitable for ionization balance calculations.



"Mechanism" – kinetic scheme with associated rate coefficients, validated over a range of conditions by comparison with expts. The detail required depends on the questions being addressed by the model.

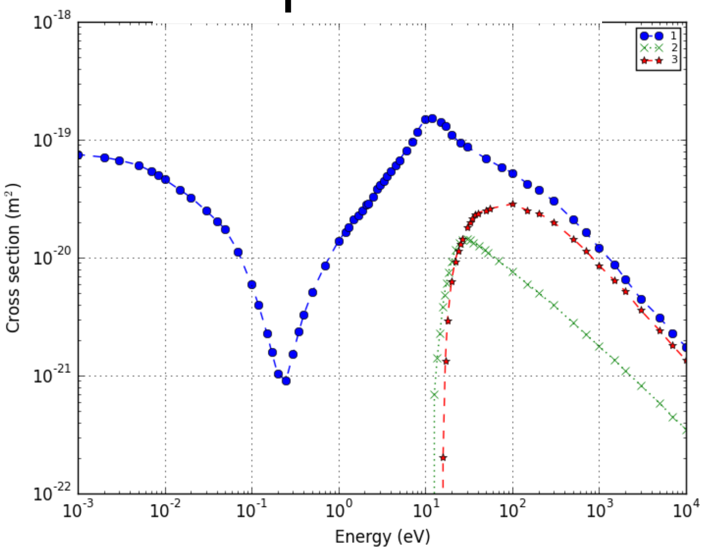
See Adamovich et al, PSST 2017, The 2017 Plasma Roadmap: LTP science & technology

LXCat policy (1)

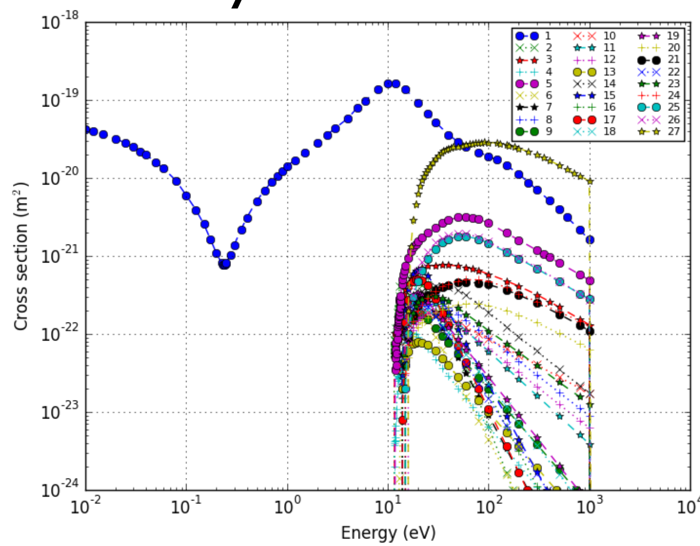
1) Anyone willing to contribute data to the site can request a password and set up a database. => Data for the same processes can be listed in multiple databases. ***LXCat does not recommend data.***

Example: complete sets of electron-Argon cross sections

Phelps database



Hayashi database



LXCat policy (2)

2. The site is ***open access*** and data can be downloaded without registering or paying a fee, but ***proper referencing is essential*** for the survival of LXCat.

Downloaded data should not become anonymous!

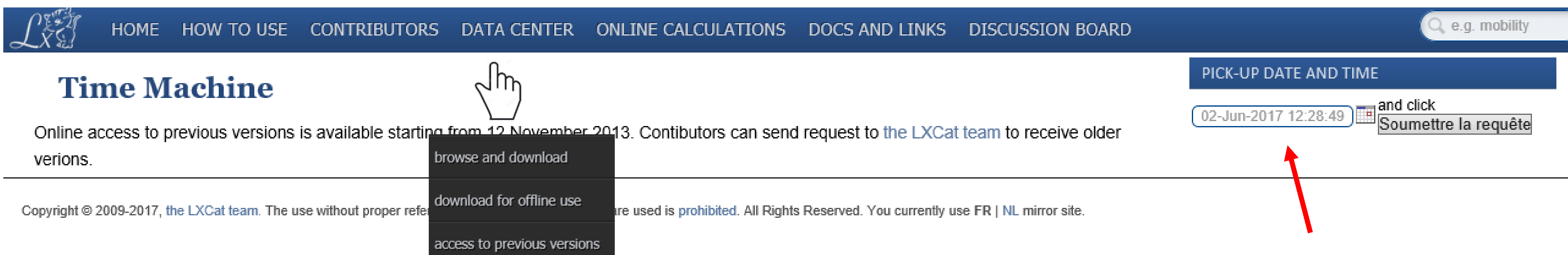
Required reference format:
[database name], www.lxcat.net, [retrieved date]

+ List all references given in the database for the species

Example: Hayashi database, www.lxcat.net, retrieved June 7, 2017
M. Hayashi (2003) "Bibliography of electron and photon cross sections with atoms and molecules published in the 20th century - argon", report NIFS-DAT-72 of the National Institute for Fusion Science of Japan

LXCat policy (3)

3. ***Databases are dynamic***. Contributors make changes as new data become available or when corrections are needed. ***Data as they existed at a specific data in the past can be recovered on-line.***



Set date to retrieve previous versions of data

Conclusions

This talk focused on the data related to the electron component in LTPs. **Data needs are still far from being satisfied.**

It is becoming very important for the LTP community to establish and distribute recommended **reaction mechanisms**, at least for for common gas mixtures. Actions underway towards this end include a round-robin exercise and a proposed COST action (EU) coordinated by Miles Turner.

Plasma surface/liquid interactions are areas of continuing/emerging interest for which data are largely unavailable.

END

What's been learned from the LXCat experience

Reasons why LXCat is a success:

- responds to a **well-recognized need**
- well-defined **scope**
- easy to use, **open-access**, on-line tools….
- a **community-wide effort**
- responsive but not beholden to **commercial activities**
- **compatible** with existing software
- **nurtured** by the GEC

Outstanding issues:

- long-term **survivability** (non-profit association)
- efficient use of limited **resources**
- maintain **visibility** of LXCat and its contributors
- **recommended data?**

lxcat.info@gmail.com