Laser
LISOL
Source

# In-Gas-Jet Resonance Ionization Spectroscopy

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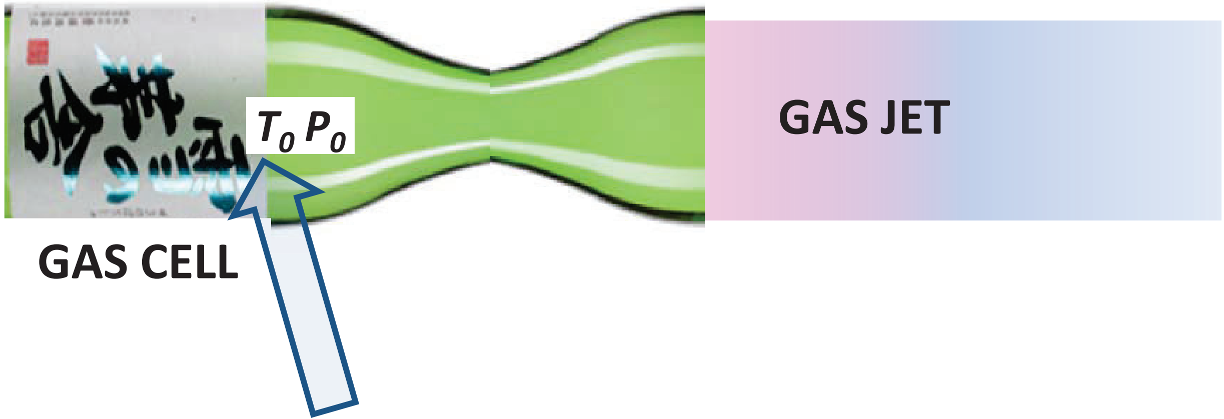
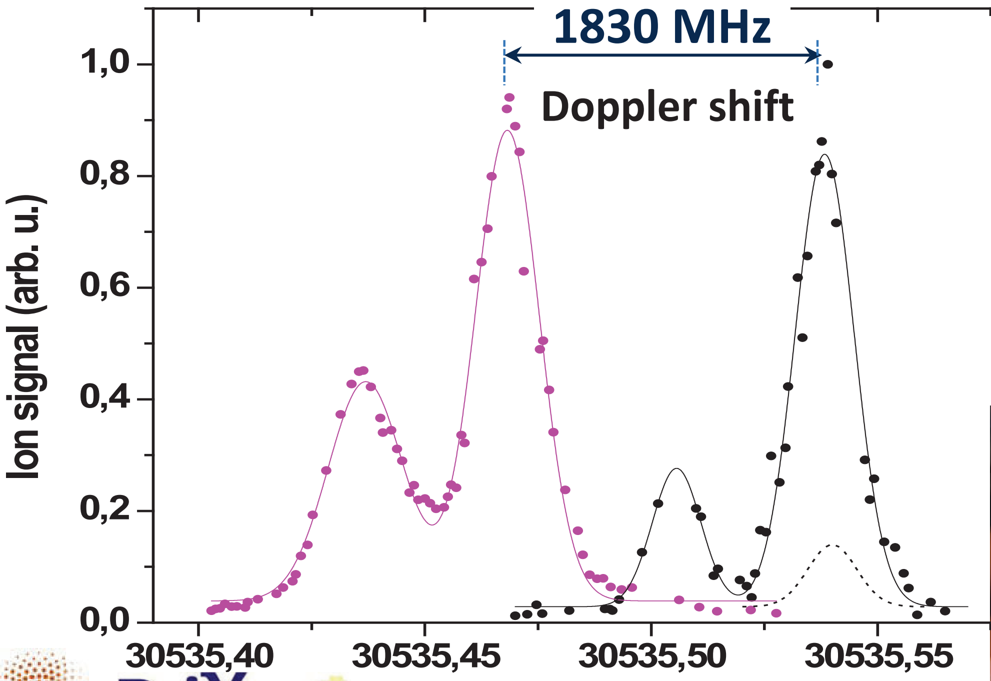
1. What spectral resolution is achievable in the gas jet RIS ? And How?
2. Requrements for laser radiation, pumping system …

# Types of nozzles that will be discussed

## Convergent- Divergent Nozzle

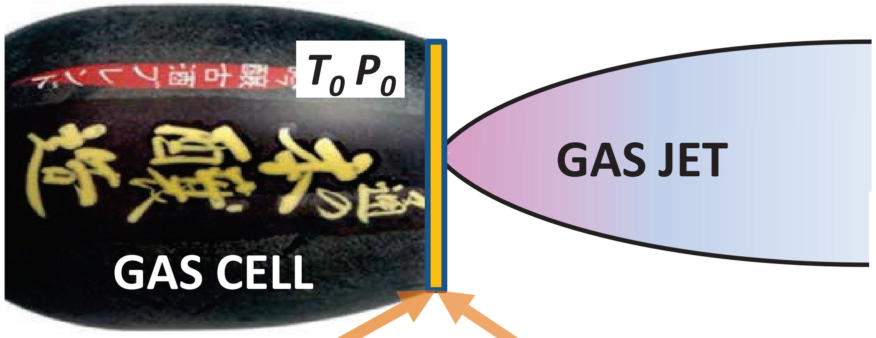


$T_0$ =355±3K

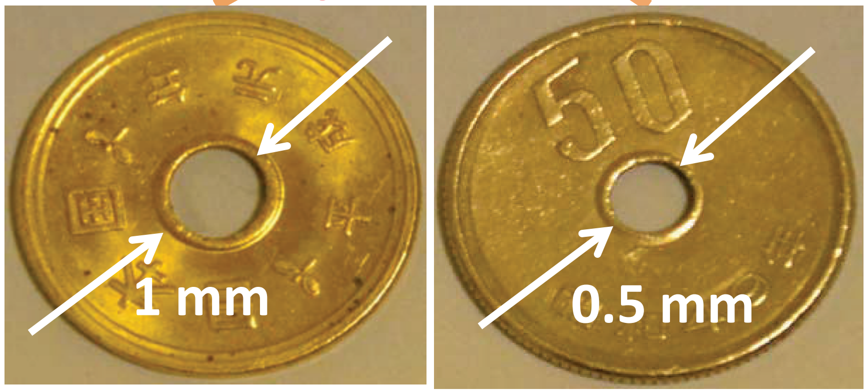


## Free-Jet Nozzle

TOP VIEW



SIDE VIEW



Exit orifice diameter

Pumping speed to obtain the desired Mach number

# Properties of supersonic beams (I)

$$CpT_0 = CpT + \frac{u^2}{2} \qquad u_{\max} = \sqrt{2CpT_0}$$

The sum of specific enthalpy and kinetic energy remains constant during expansion

**Mach number** - $\boxed{M = u/a}$

$\boxed{a = \sqrt{\frac{\gamma kT}{m_{ng}}}}$

***u*** **- gas stream velocity**

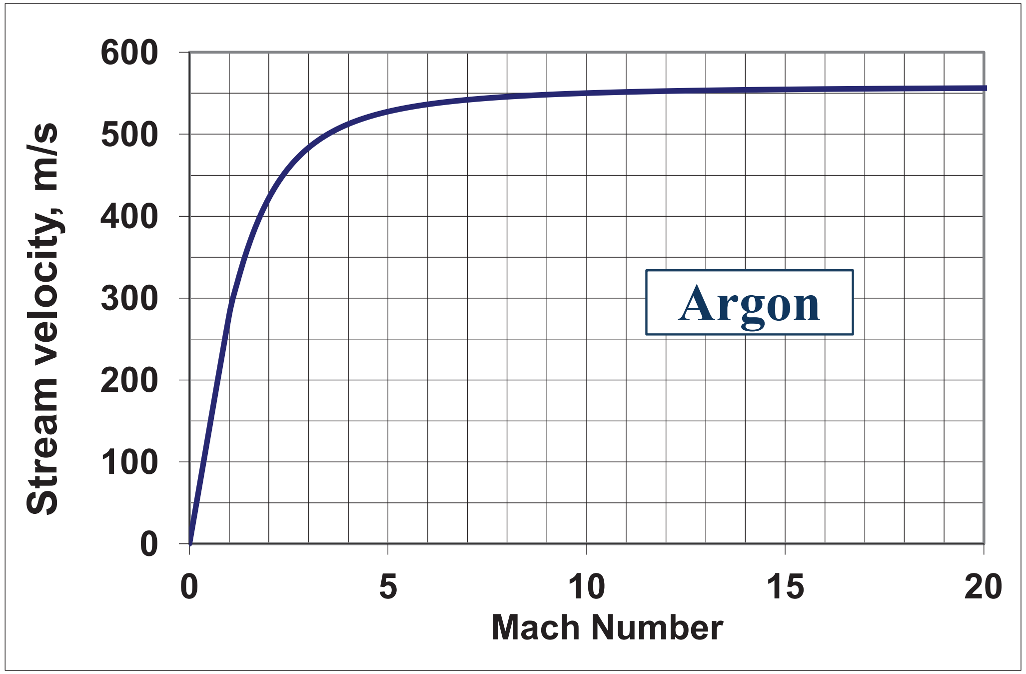
***a*** **- local speed of sound**

***T*** **- gas temperature**

***mng*** **- mass of the noble gas**

$$\boxed{u = \sqrt{\frac{kT_0\gamma M^2}{m_{ng}\left\{1+\left[(\gamma-1)/2\right]M^2\right\}}}}$$

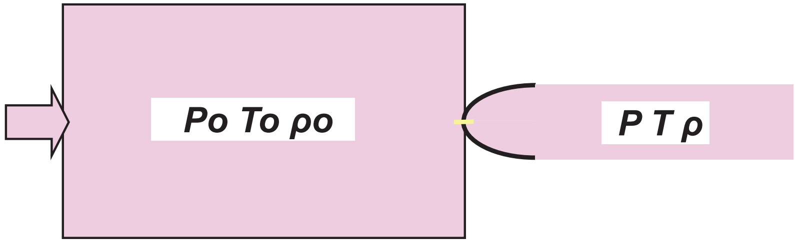
$\gamma = Cp/Cv$ - ratio of specific heat capacities =5/3



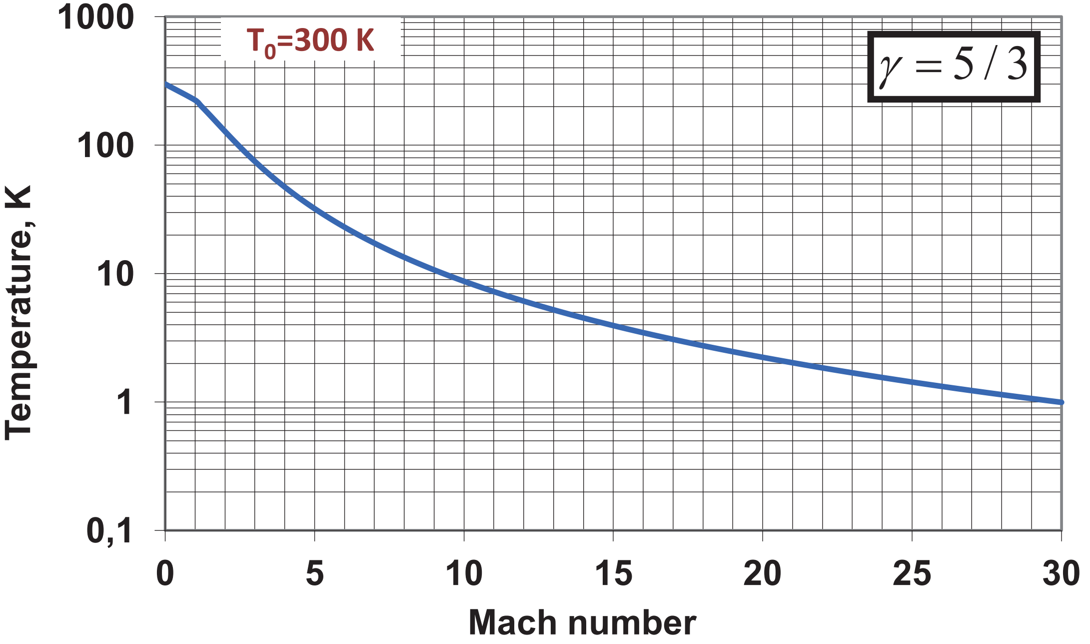
**Stream velocity Ar-550 m/s**

**He ~ sqrt $(m_{ng})^{-1}$**

# Properties of supersonic beams (II)



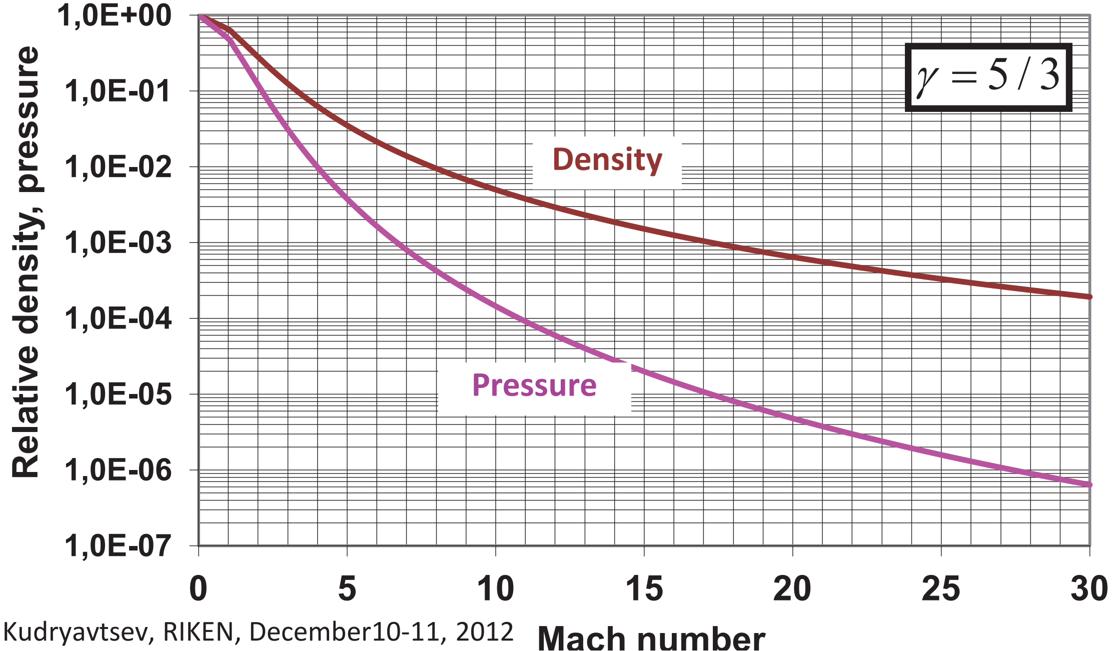
Isentropic flow - gas entropy is constant



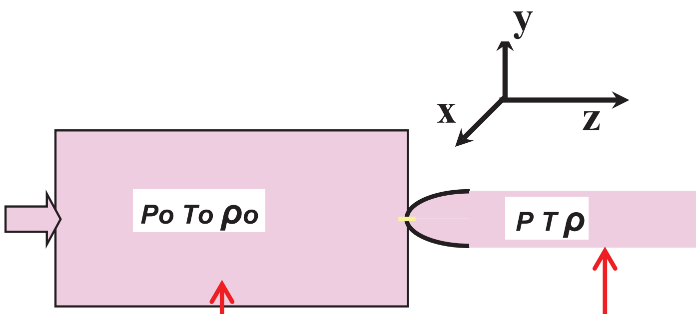
$$\frac{T}{T_0} = \left[1 + \left(\frac{\gamma - 1}{2}\right) M^2\right]^{-1}$$

$$\frac{\rho}{\rho_0} = \left[1 + \left(\frac{\gamma - 1}{2}\right) M^2\right]^{\frac{-1}{\gamma - 1}}$$

$$\frac{P}{P_0} = \left[1 + \left(\frac{\gamma - 1}{2}\right) M^2\right]^{\frac{-\gamma}{\gamma - 1}}$$



# Properties of supersonic beams (III)

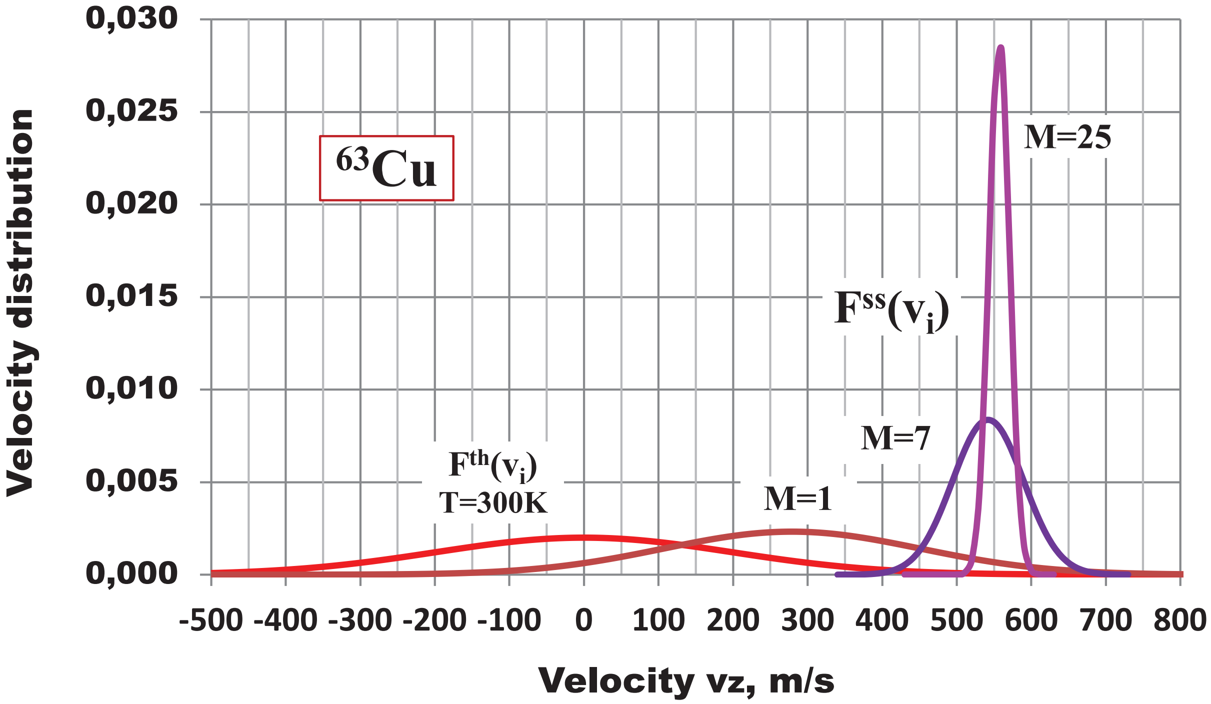


$$F^{th}(v_i) = \sqrt{\frac{m}{2\pi k T_0}}\,\exp\left(\frac{-m v_i^2}{2kT_0}\right)$$

$$F^{ss}(v_z) = \sqrt{\frac{m}{2\pi k T}}\,\exp\left(\frac{-m(v_z-u)^2}{2kT}\right)$$

**Radioactive atoms are in thermal equilibrium with buffer gas atoms**

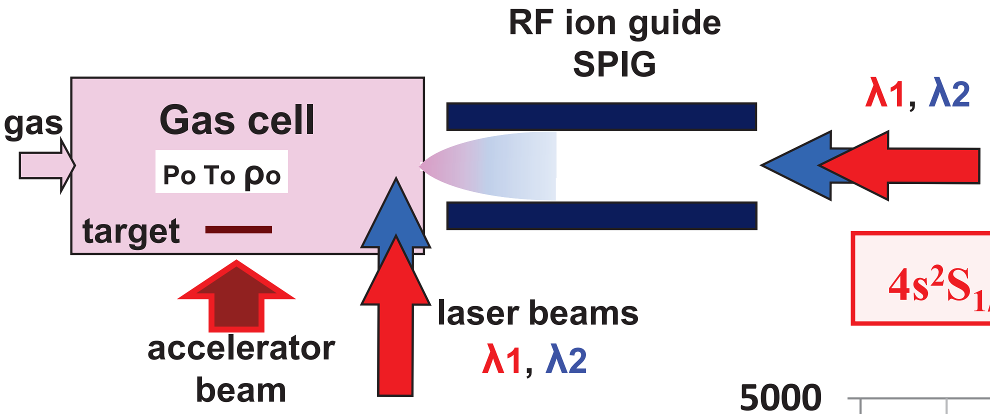
**One dimensional Maxwell-Boltzmann velocity distribution**



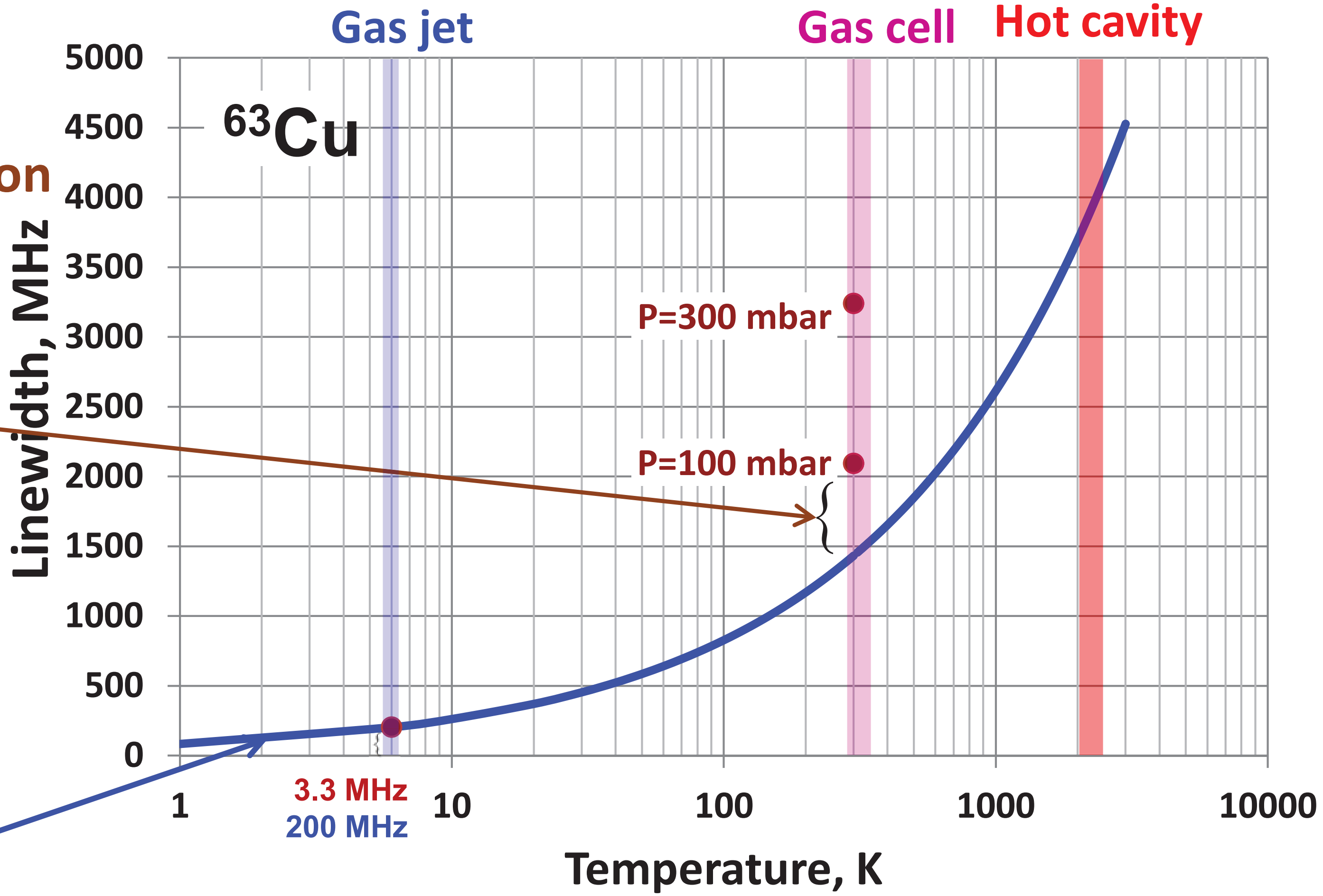
$$\delta(F) = 2\sqrt{\ln 2}\sqrt{\frac{2kT}{m}}$$

**- Full Width at Half Maximum (FWHM)**

# Doppler and Collision Contributions to the Spectral Line Width



$4s^2S_{1/2} – 4p^2P_{1/2}$, 327.4 nm $^{63}$Cu transition, $\nu_0$= 30535.3 cm$^{-1}$



## Collision/pressure contribution

$$\Delta\nu_{coll} = \gamma_{coll} \times \rho$$

$\gamma_{coll}$ - collision broadening coefficient, 1.5·10$^{-20}$ cm$^{-1}$/cm$^{-3}$ (8 MHz/mbar)

$\rho$ – gas density (atom /cm$^3$)

## Doppler contribution

$$\Delta\nu_{Doppler} = 2\sqrt{\ln 2}\,\frac{\nu_0}{c}\sqrt{\frac{2kT}{m}}$$

Yu. Kudryavtsev, EMIS2012, December 5, 2012

# Doppler Gaussian and collision- and natural Lorentzian contributions to the spectral line shape

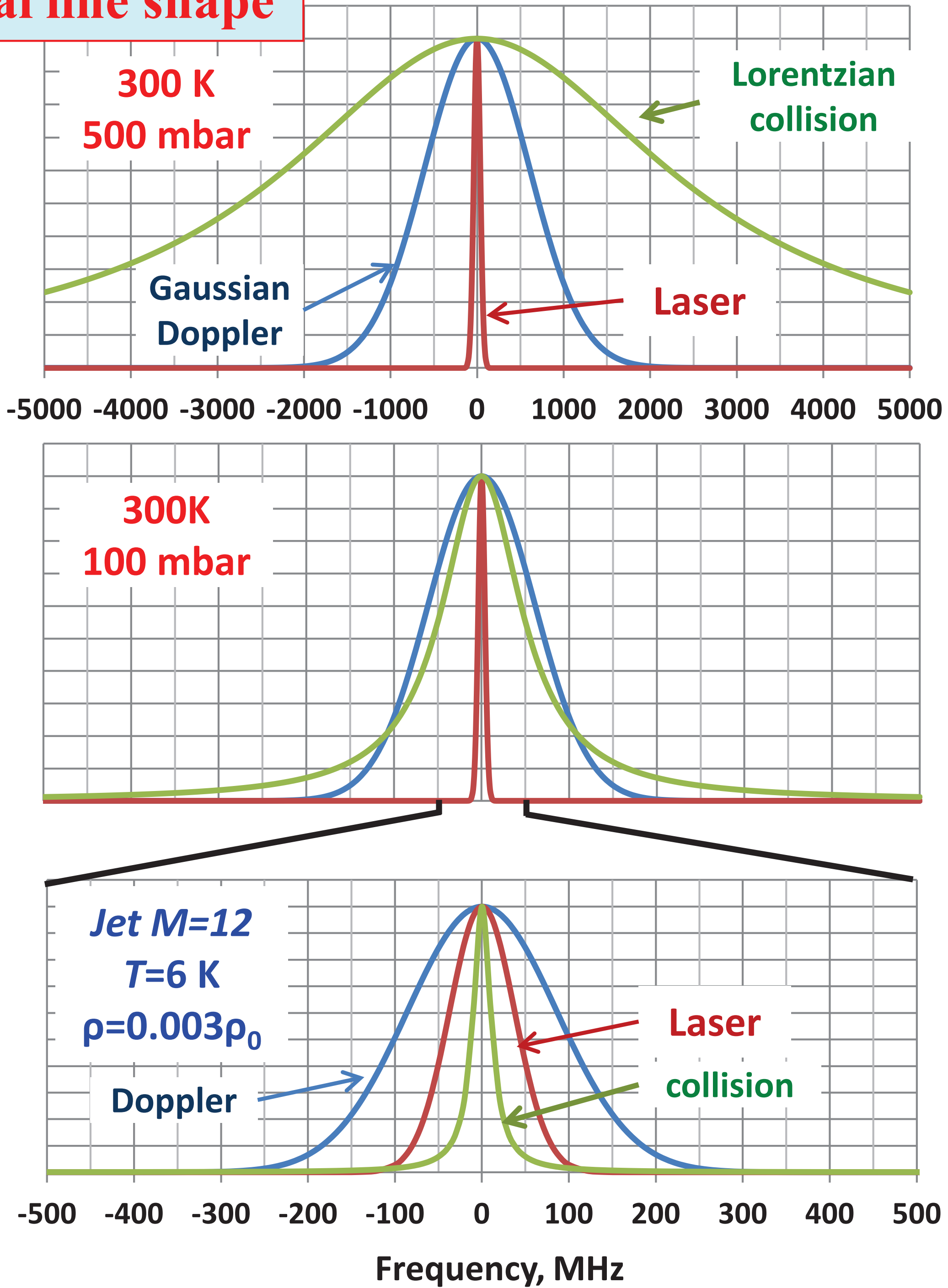
$$G(\nu) = G_0 \exp\left[ -\frac{c^2(\nu-\nu_o)^2}{\nu_0^2 \frac{2kT}{m}} \right]$$

$$L(\nu-\nu_0) = \frac{1}{2\pi}\frac{\Gamma}{(\nu-\nu_0+\Gamma sh)^2+(\Gamma/2)^2}$$

Laser bandwidth – $\delta_{laser}$ Gaussian if laser time profile is Gaussian

$$\boxed{\delta_{laser} = 441/\tau_{pulse}}$$

$\tau_{pulse} = 5$ ns $\delta_{laser}$ =88 MHz



# Laser Resonance Ionization in Supersonic Beams

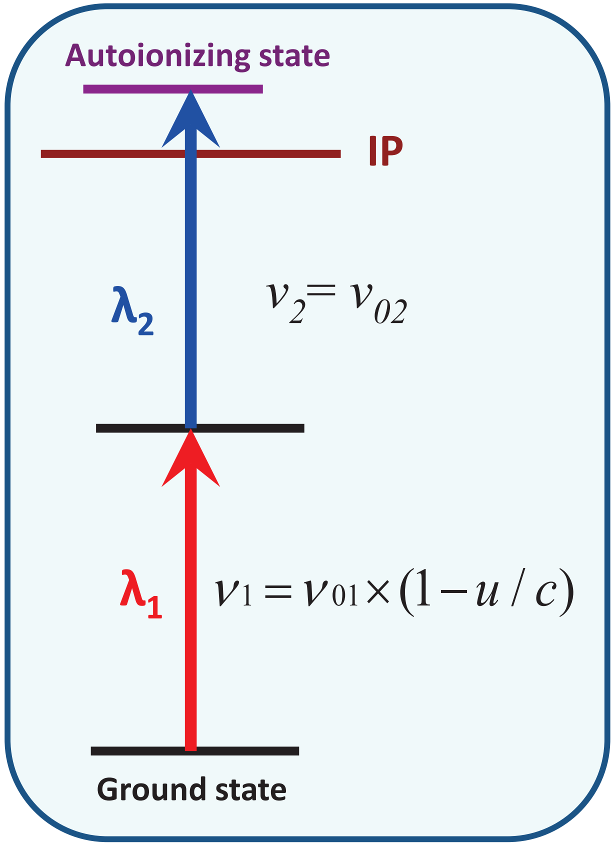
**NO laser ionization inside the cell !**

**Laser ionization- only in the cold jet**

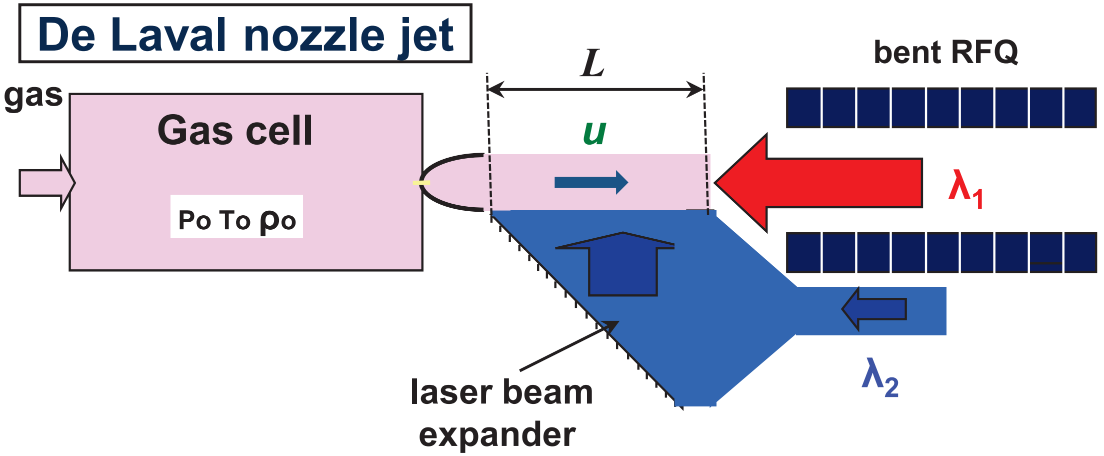
**λ$_2$ !**

$f_{laser} \geq 1/(L/u)$ **≥ 10 kHz, argon jet - L = 5.5 cm**

$u$ – **stream velocity, 550m/s**



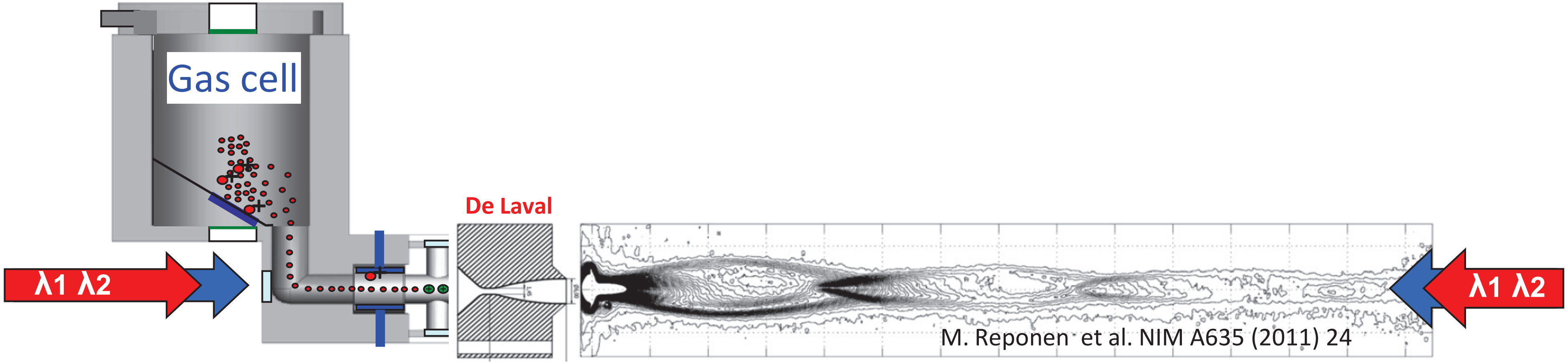
## Crossed laser beams with supersonic jet



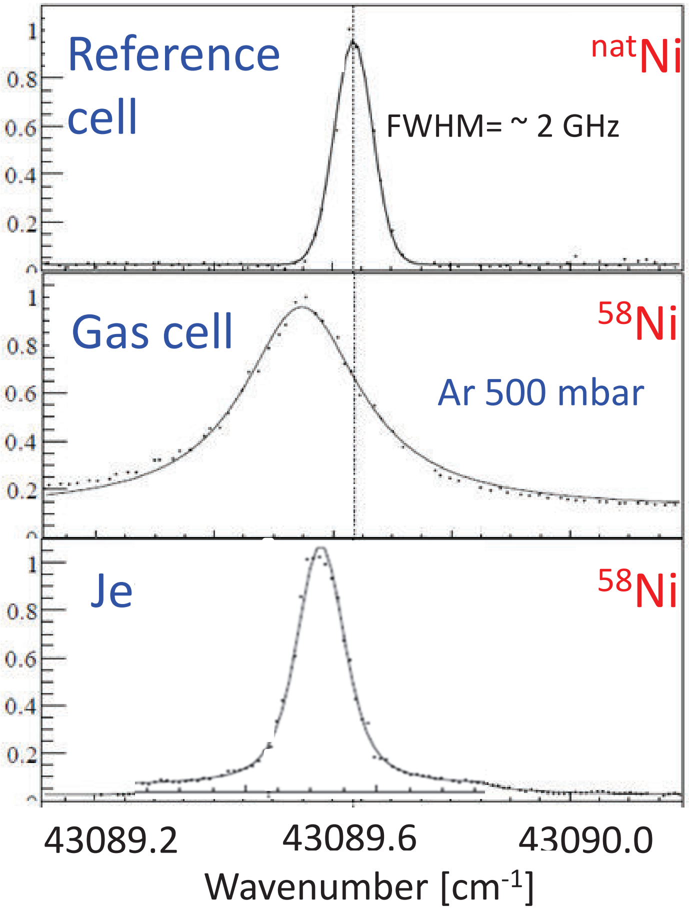
**The parallel beam from de Laval nozzle !**

**No broadening due to the beam divergence**

**Very careful design of the nozzle is required**



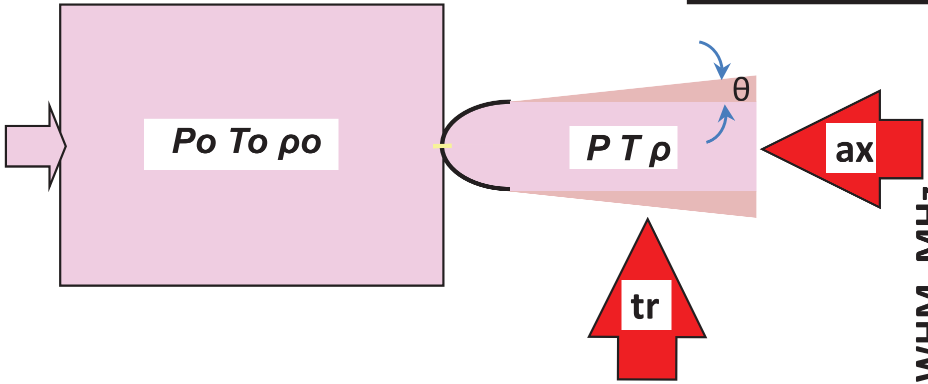
M. Reponen et al. NIM A635 (2011) 24



T. Sonoda et al. NIM B267 (2009) 2908

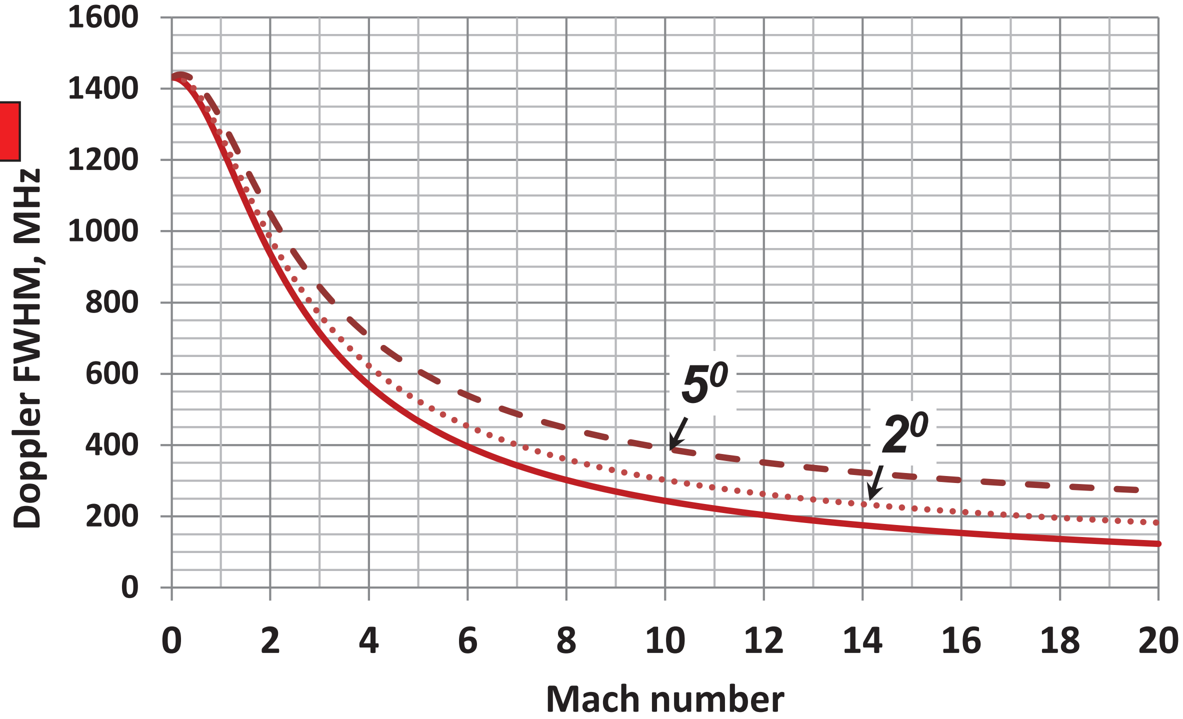
1. **Contra-propagating direction of laser- and supersonic beams makes conditions even worth in comparison co-propagating one**
2. **Applying blocking potential to the RF ion guide will lead to collection of laser-produced ions**
3. **No ionization in shock wave zones**

# Doppler broadening in supersonic beam



$$\Delta\nu_{Doppler} = 2\sqrt{\ln 2}\,\frac{\nu_0}{c}\sqrt{\frac{2kT}{m}}$$ - *FWHM*

$4s^2S_{1/2} – 4p^2P_{1/2}$, 327.4 nm $^{63}$Cu transition, $\nu_0$= 30535.3 cm$^{-1}$



$$\frac{A}{A^*} = \frac{1}{M}\left[\left(\frac{2}{\gamma+1}\right)\left(1+\frac{\gamma-1}{2}M^2\right)\right]^{(\gamma+1)/2(\gamma-1)}$$

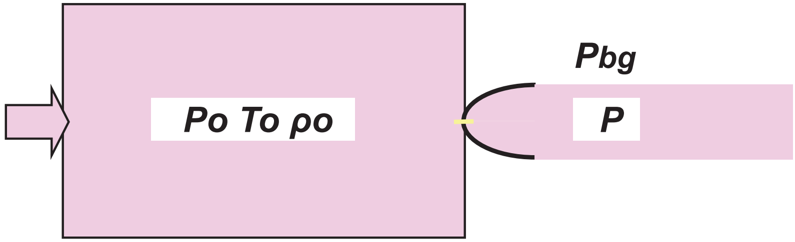
**$A^*=\pi d^2/4$**

$d$=1 mm

$M$ =7  Diameter- 4.88 mm

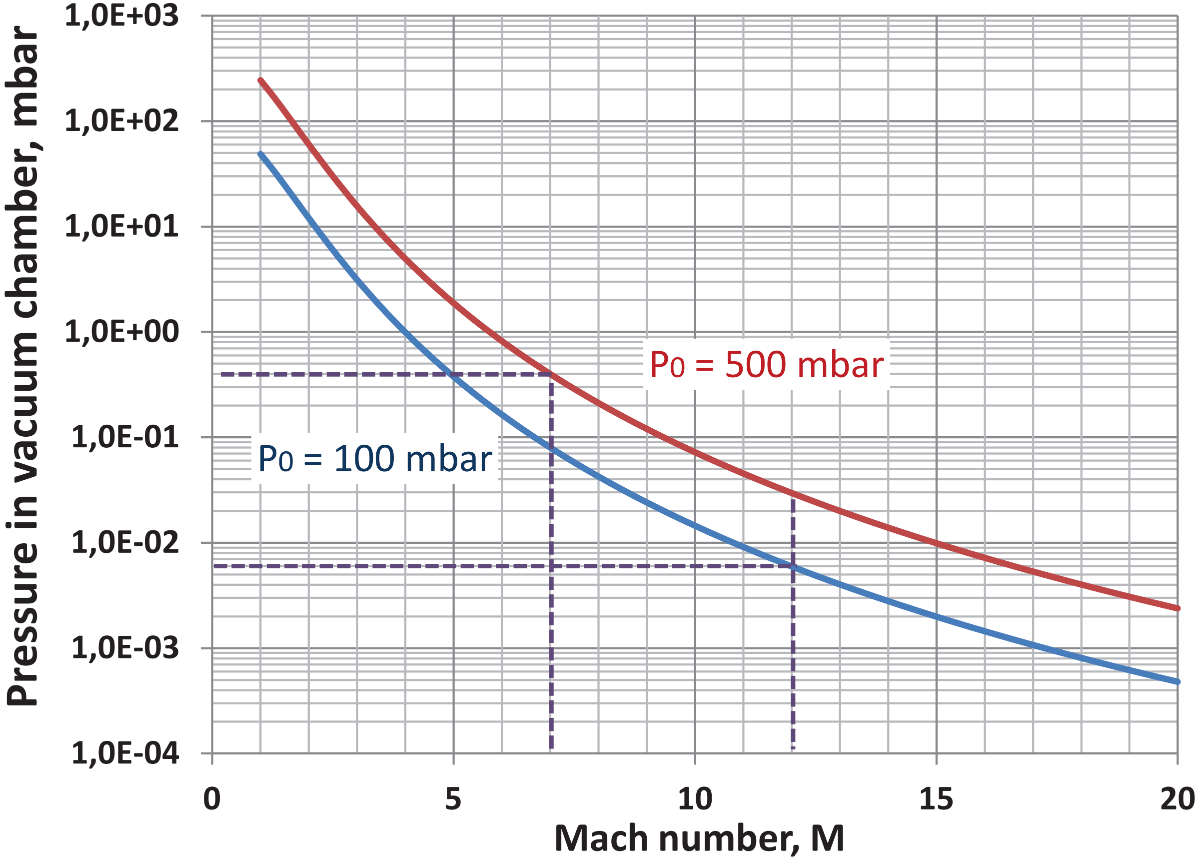
$M$ =12 Diameter - 10.51mm

# Required pumping capacity for the vacuum system (I)



$$\boxed{P_{bg} = P} \quad !$$

$$\frac{P}{P_0} = \left[1 + \left(\frac{\gamma - 1}{2}\right) M^2\right]^{\frac{-\gamma}{\gamma - 1}}$$



# Required pumping capacity for the vacuum system (II)

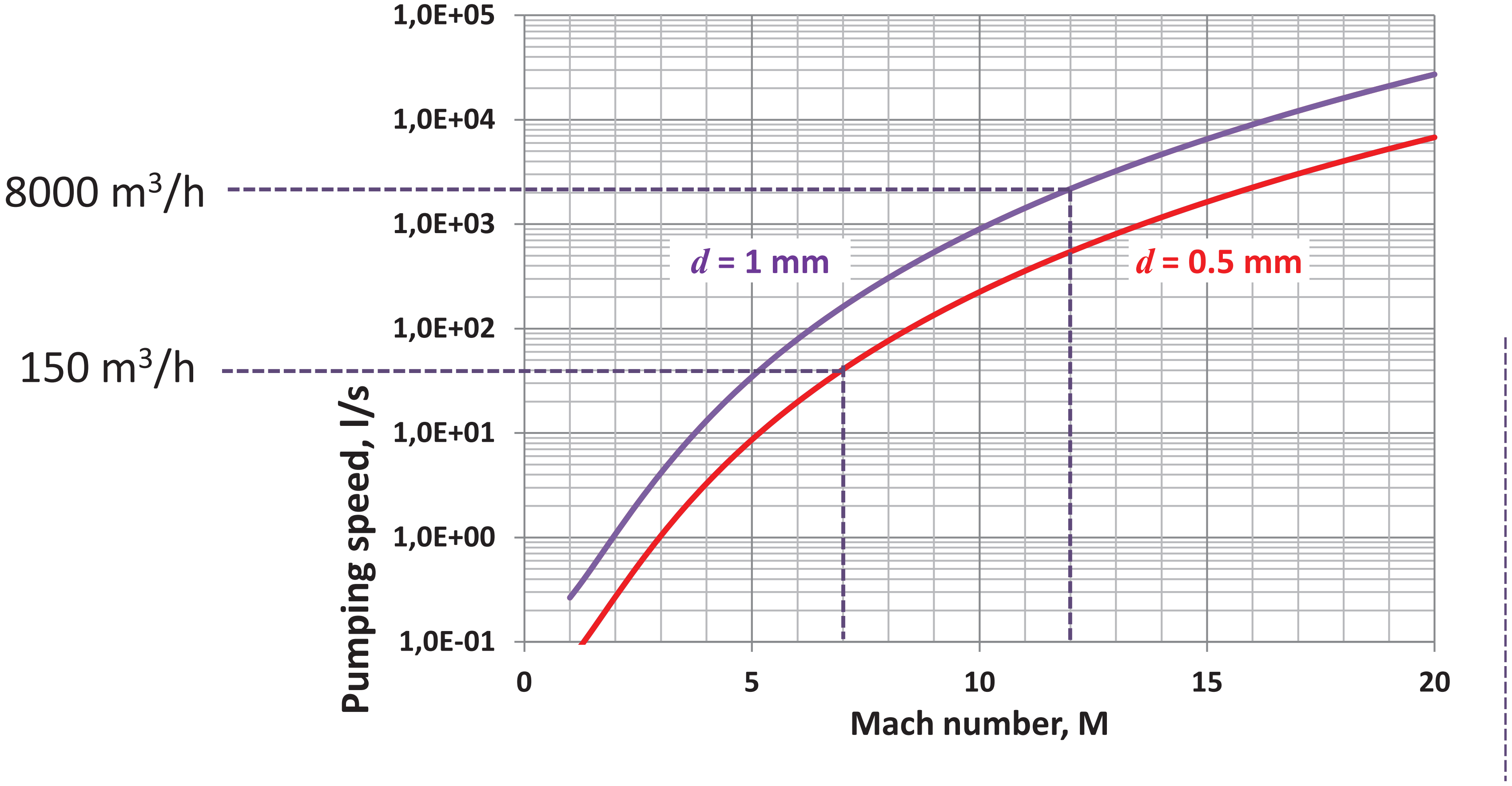
$Q$ - the volume flow rate of the buffer gas through the throat diameter $d$ (in l/s)

$$Q = 0.052 d^2 \sqrt{\frac{T_0}{A}}$$

$A$ - in atomic mass units

$W$ - pumping speed of the vacuum system

$$W = \frac{QP_0}{P_{bg}} \qquad \boxed{P_{bg} = P}$$



# Formation of clusters and chemical reactions

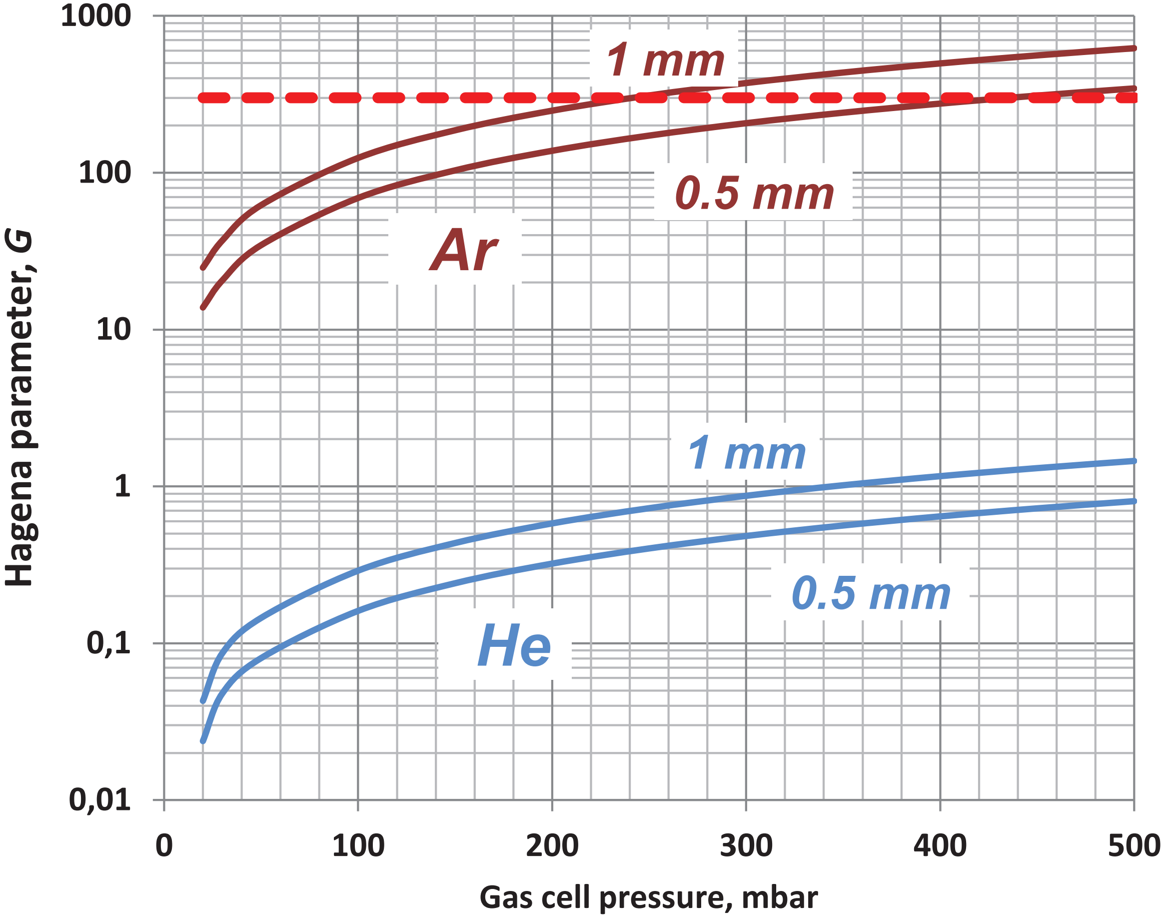
Hagena parameter G*

$$G^* = \eta \frac{(d)^{0.85}}{T_0^{2.29}} P_0$$

$\eta$ is the condensation parameter related to the bond formation.

number of atoms per clusters
$Nc \sim G^{*2.0\text{-}2.5}$

$Nc \sim T_0^{-5}$

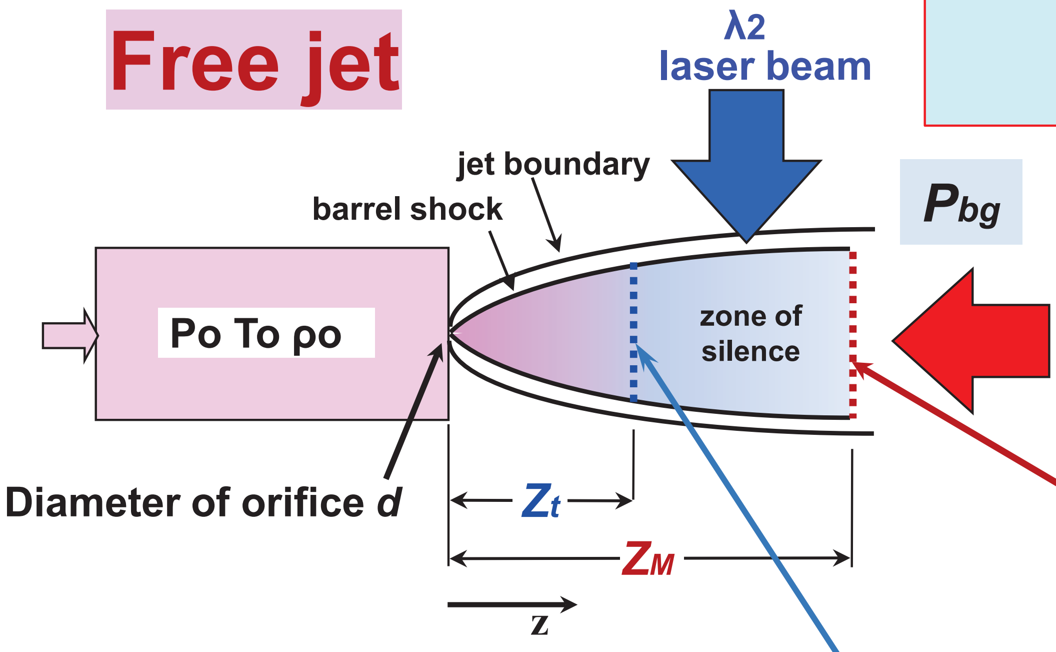


Clustering starts if the Hagena parameter $G^* > 300$

# Two-Step Laser Ionization in a Free Jet

**1951 free jet – A. Kantrowitz, J. Grey**

## Free jet



**Mach disk, T, ρ ↑**

**$Z_M$ – position of the Mach disk**

$$\frac{Z_M}{d} = 0.67\sqrt{\frac{P_0}{P_{bg}}}$$

**$M_t$ - terminal Mach number**

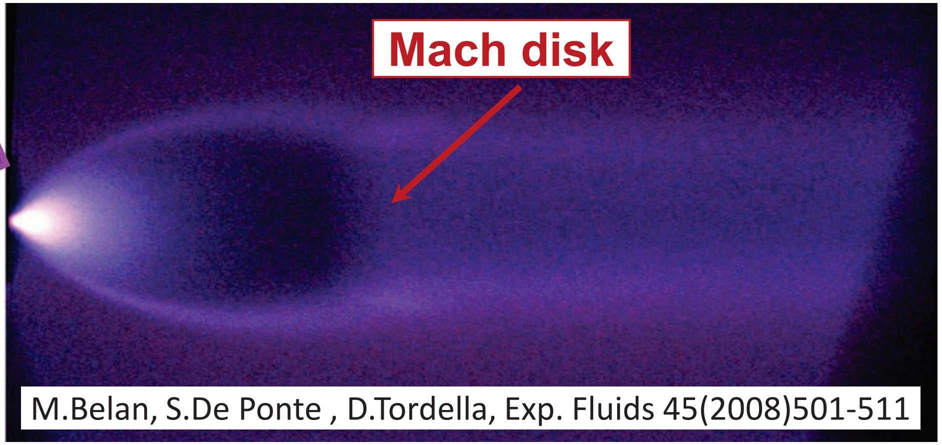
**$Z_t$ – position of terminal Mach number**

$$M_t = 3.32\left(P_0 d\right)^{0.4}$$

**(mbar, mm)**

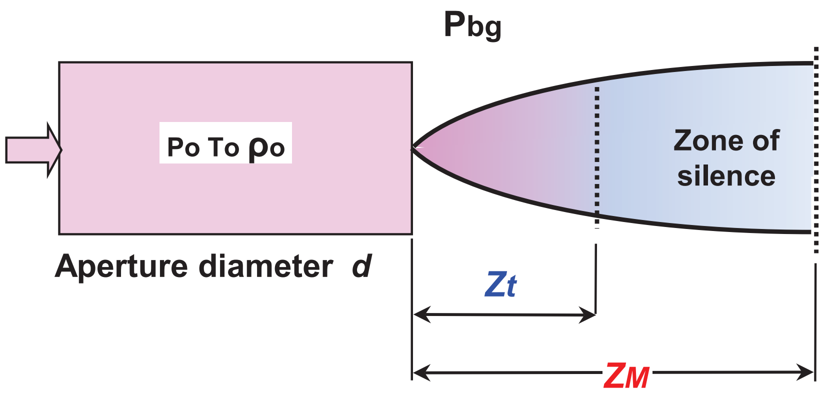
$$\frac{Z_t}{d} = \left(\frac{M_t}{3.26}\right)^{1.5}$$

## Visualization of free jet

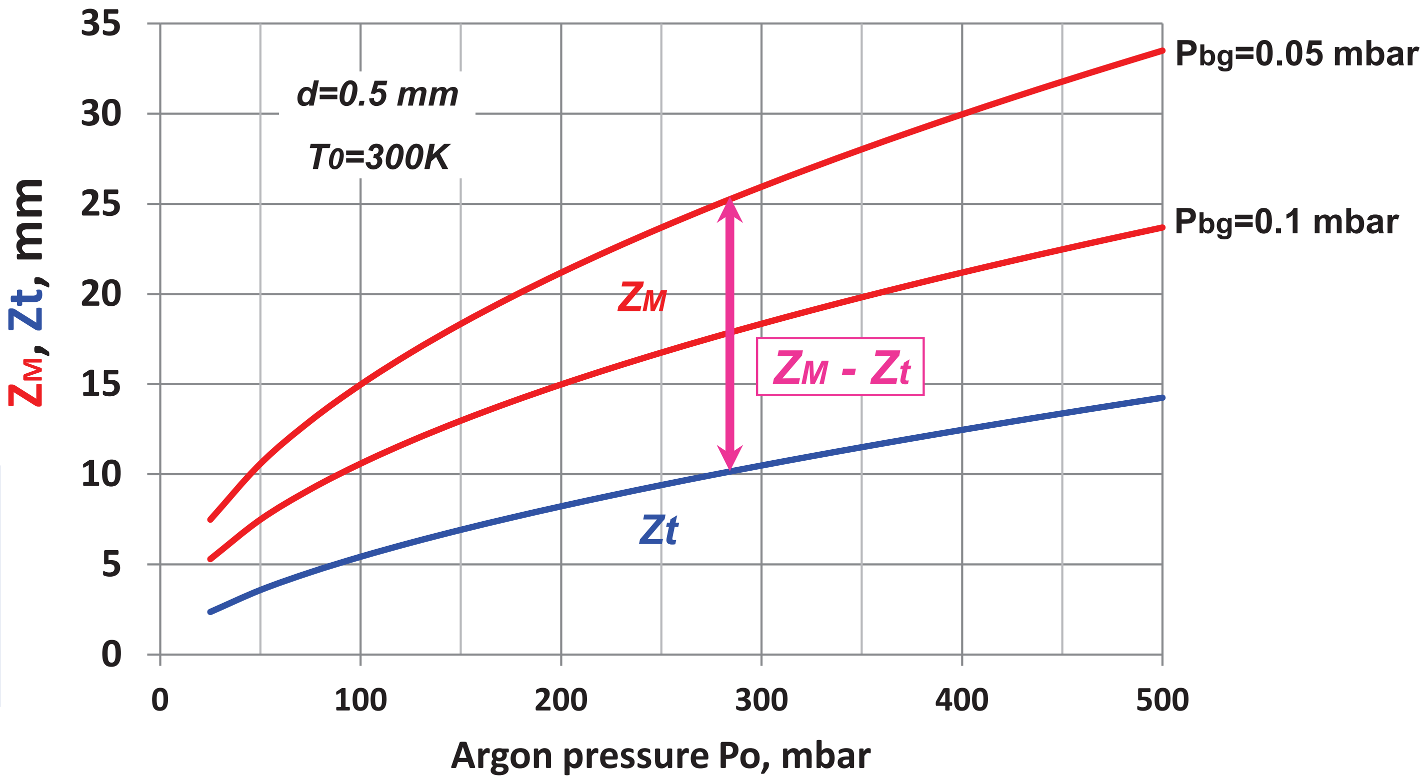


M.Belan, S.De Ponte , D.Tordella, Exp. Fluids 45(2008)501-511

# Properties of free jet (I)



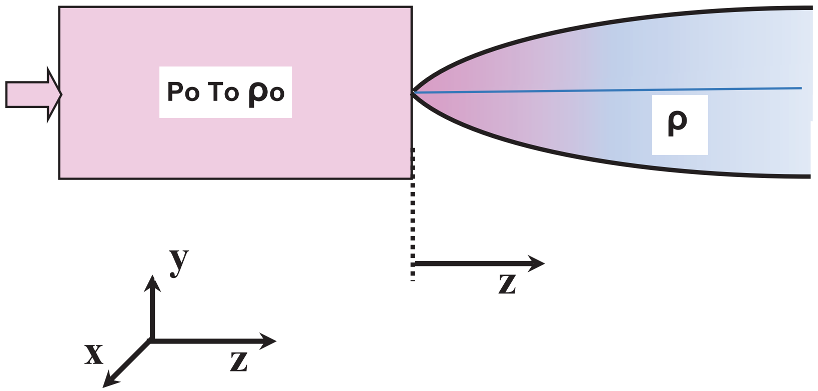
$$\frac{Z_M}{d} = 0.67\sqrt{\frac{P_0}{P_{bg}}}$$



$$\frac{Z_t}{d} = \left[\frac{133\left(P_0 d\right)^{0.4}}{3.26}\right]^{1.5}$$

Diameter of the Mach disk – $0.5 \cdot Z_M$

# Properties of Free Jet (II)

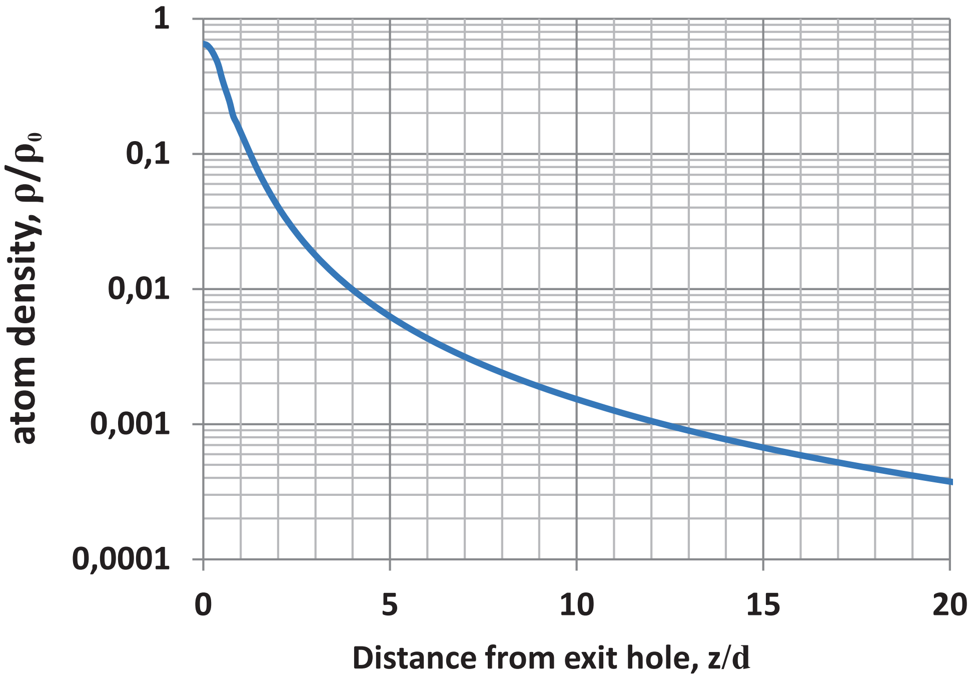


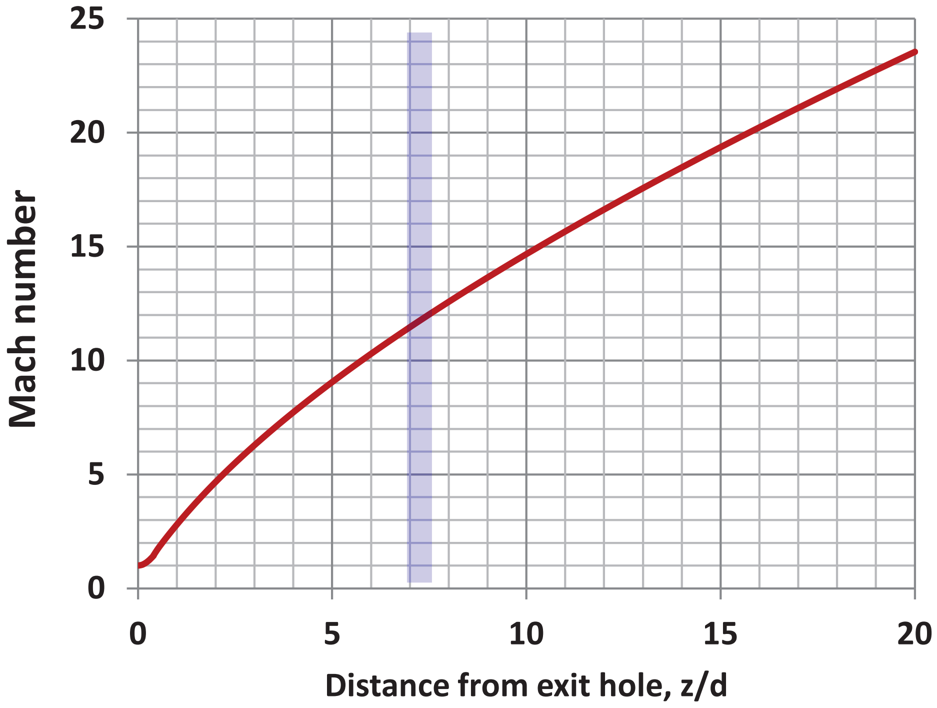
## Centerline Mach number correlations

| $A$ | $B$ | $C_1$ | $C_2$ | $C_3$ | $C_4$ |
|---|---|---|---|---|---|
| 3.337 | -1.541 | 3.232 | -0.7563 | 0.3937 | -0.0729 |

$$0<\frac{Z}{d}<1.0 \qquad M=1.0+A\left(\frac{Z}{d}\right)^{-2}+B\left(\frac{Z}{d}\right)^{3}$$

$$\frac{Z}{d}>0.5 \qquad M=\left(\frac{Z}{d}\right)^{(\gamma-1)}\left[C_1+\frac{C_2}{\left(\frac{Z}{d}\right)}+\frac{C_3}{\left(\frac{Z}{d}\right)^2}+\frac{C_4}{\left(\frac{Z}{d}\right)^3}\right]$$



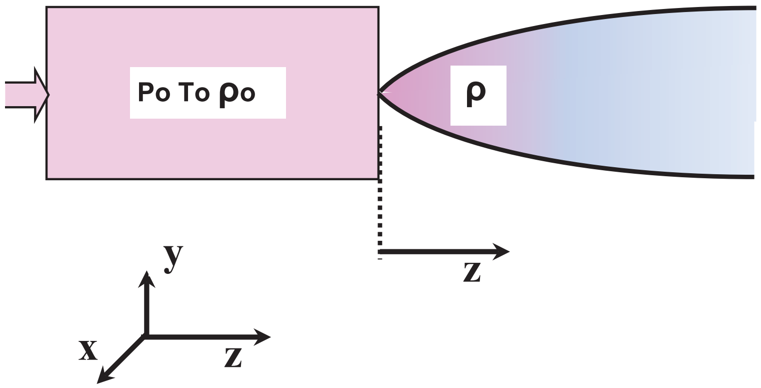


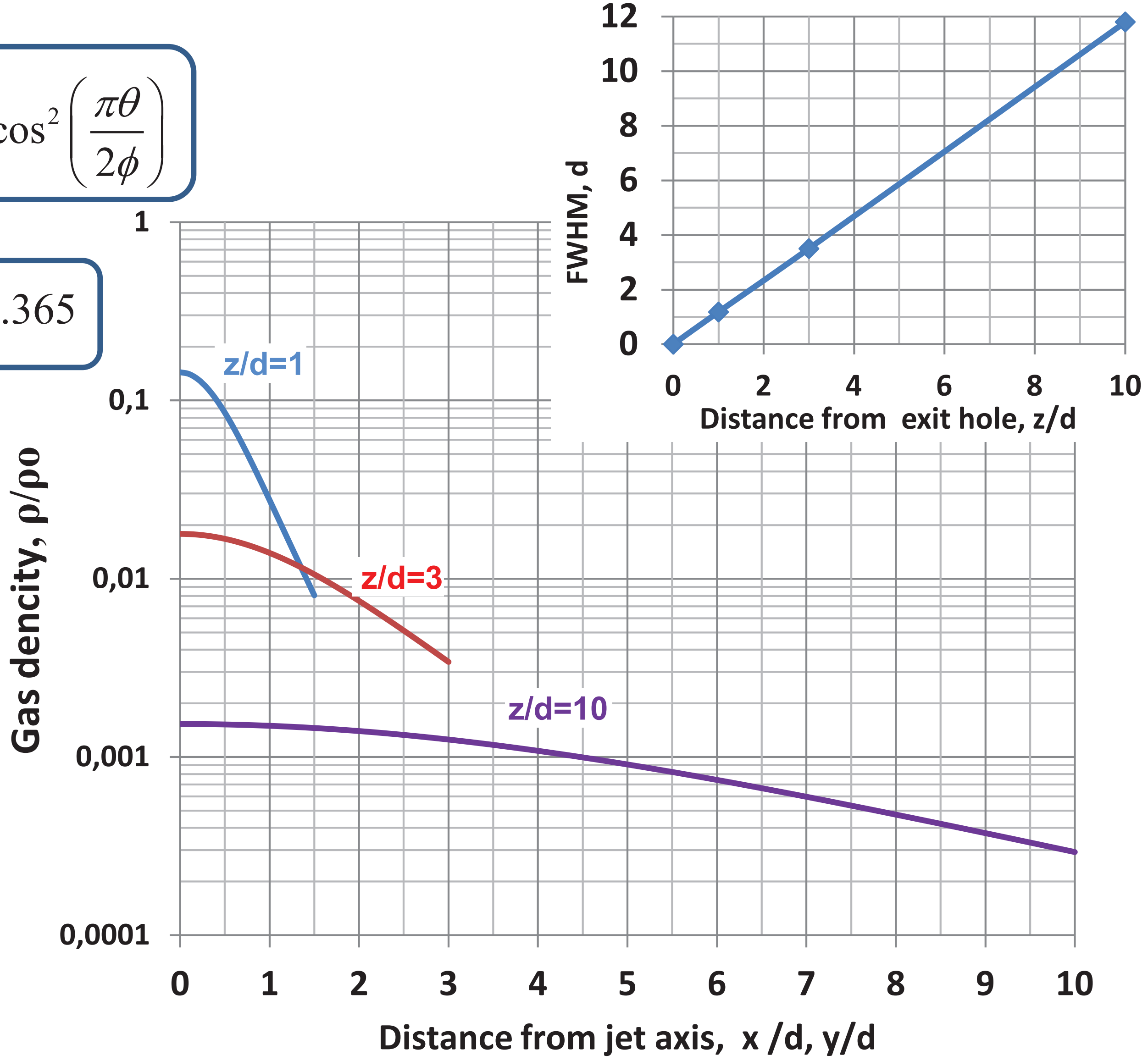
# Properties of Free jet (III)

## Off-axis density distribution for axisymmetric free jet flow

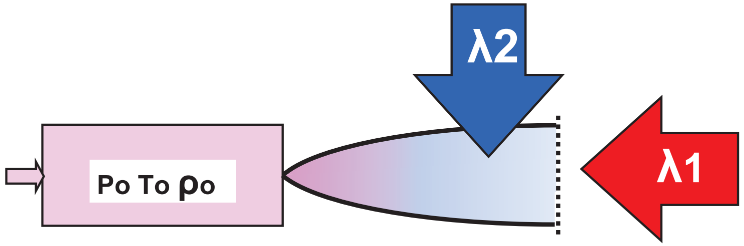
$$\frac{\rho(y,z)}{\rho(0,z)} = \cos^2\theta\cos^2\left(\frac{\pi\theta}{2\phi}\right) \qquad \frac{\rho(R,\theta)}{\rho(R,0)} = \cos^2\left(\frac{\pi\theta}{2\phi}\right)$$

$$\tan\theta = \frac{y}{z} \qquad R^2 = z^2 + y^2 \qquad \gamma = 5/3 \qquad \phi = 1.365$$

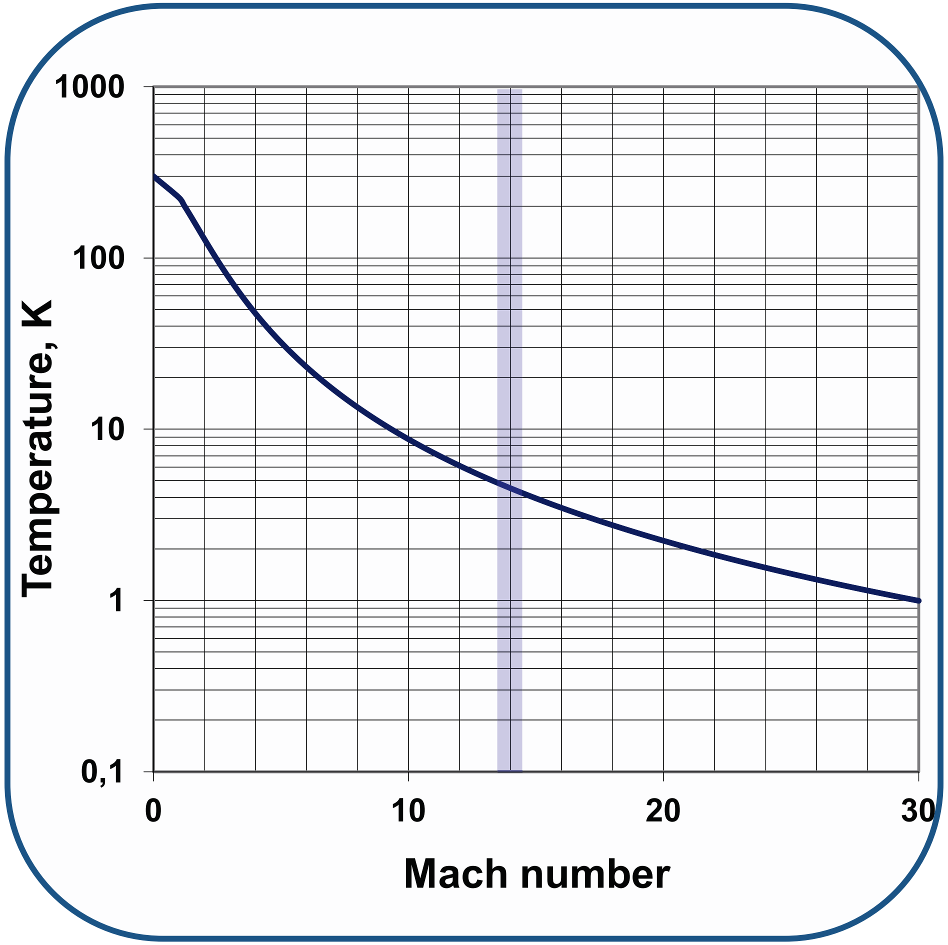


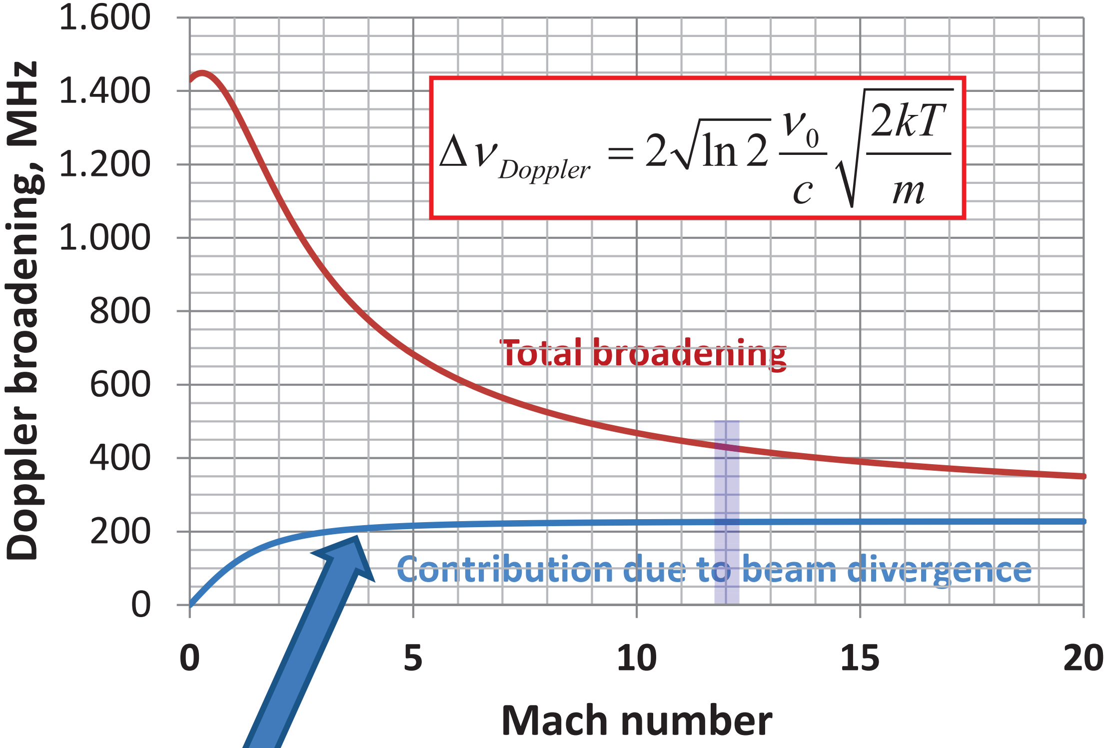


# Doppler Broadening in the Free Jet Supersonic Beam



4s$^2$S$_{1/2}$ – 4p$^2$P$_{1/2}$, 327.4 nm $^{63}$Cu transition, $\nu_0$= 30535.3 cm$^{-1}$





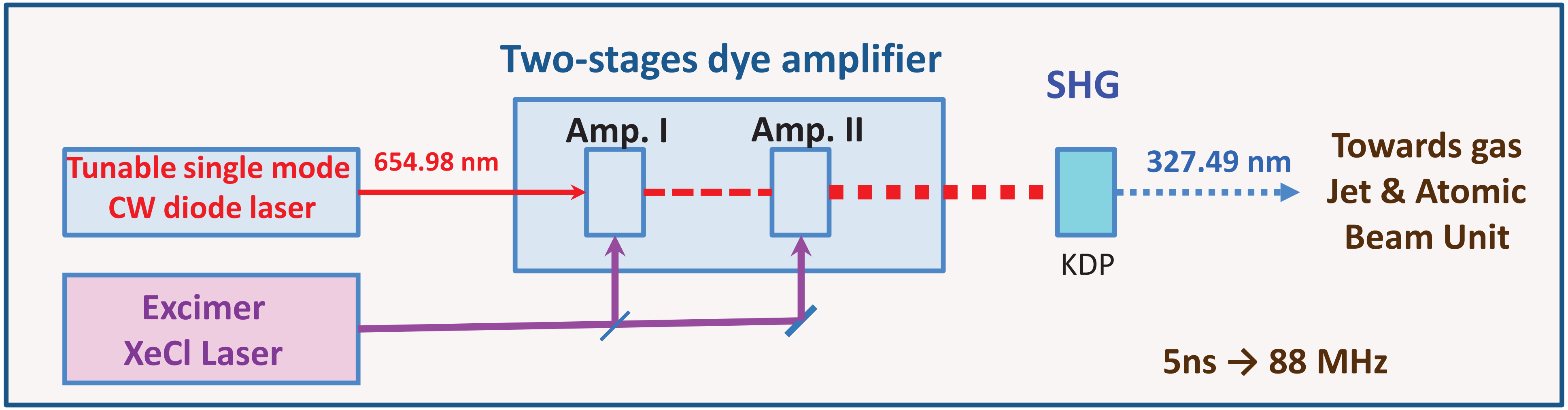
$$\Delta \nu_{Doppler} = 2\sqrt{\ln 2}\,\frac{\nu_0}{c}\sqrt{\frac{2kT}{m}}$$

$\Delta^{ax}_{Doppler} = \nu_0 \cdot u\left(1-\cos\theta\right)/c$ - axial laser beam direction

**T=4K, Dopp. W.=200 MHz**

**Tot. broad. = 420 MHz**

# Amplification of CW Single Mode Diode Laser Radiation in a Pulsed Dye Amplifier



Two-stages dye amplifier

Amp. I

Amp. II

SHG

Tunable single mode CW diode laser

654.98 nm

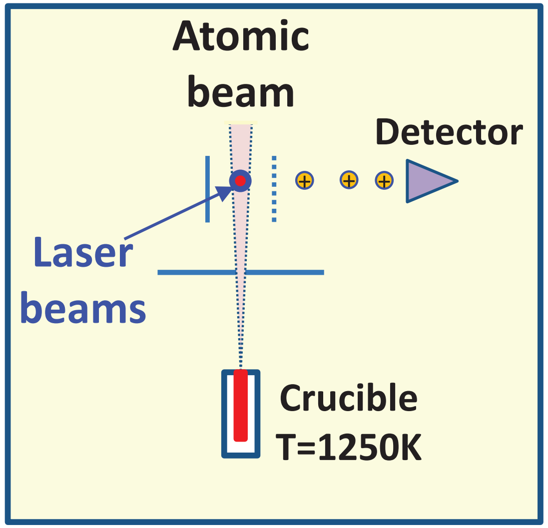
327.49 nm

KDP

Towards gas Jet & Atomic Beam Unit

Excimer XeCl Laser

5ns → 88 MHz

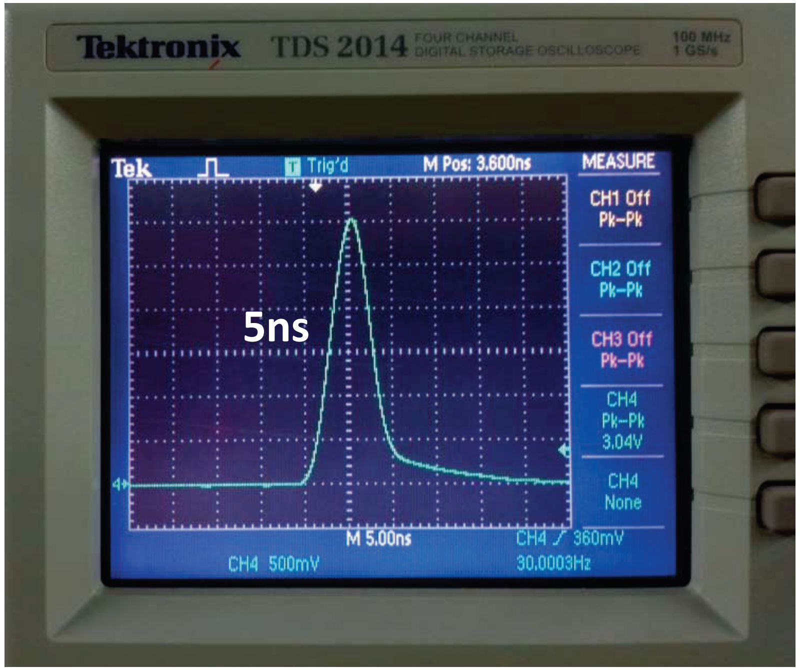


Atomic beam

Detector

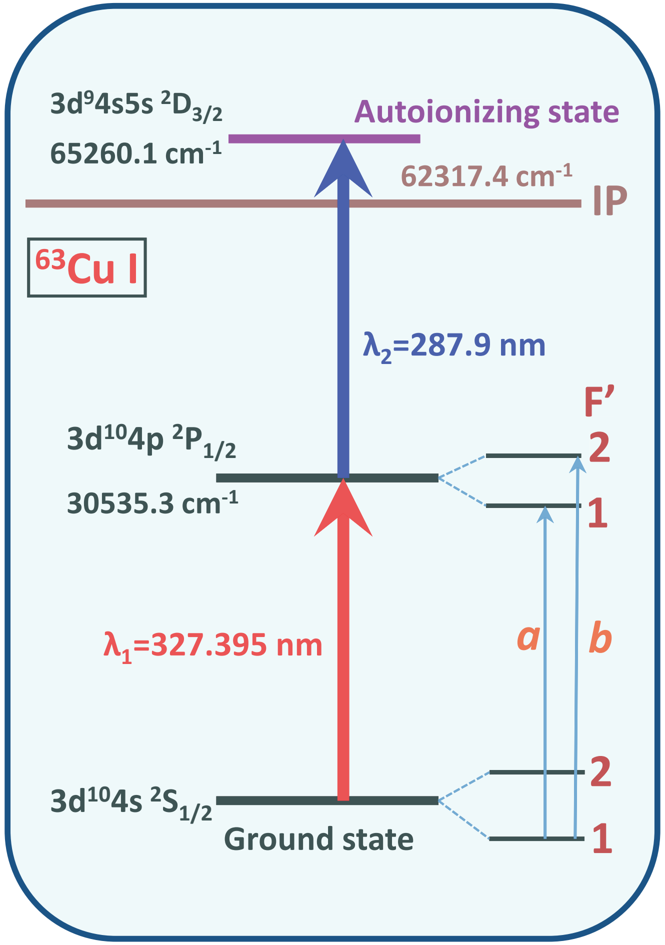
Laser beams

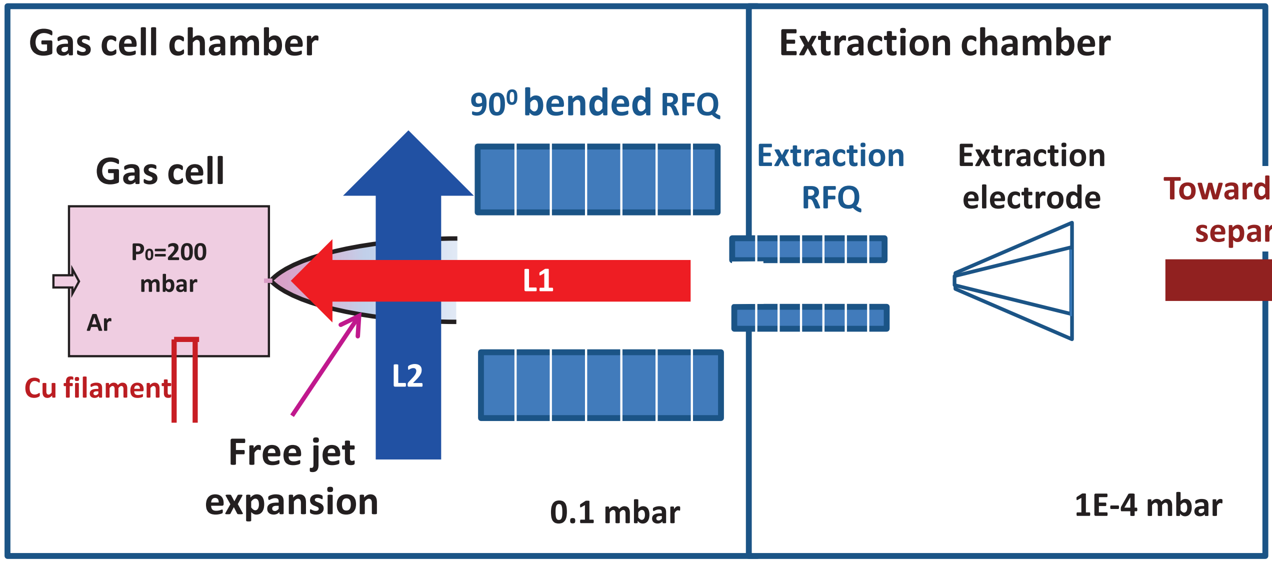
Crucible T=1250K

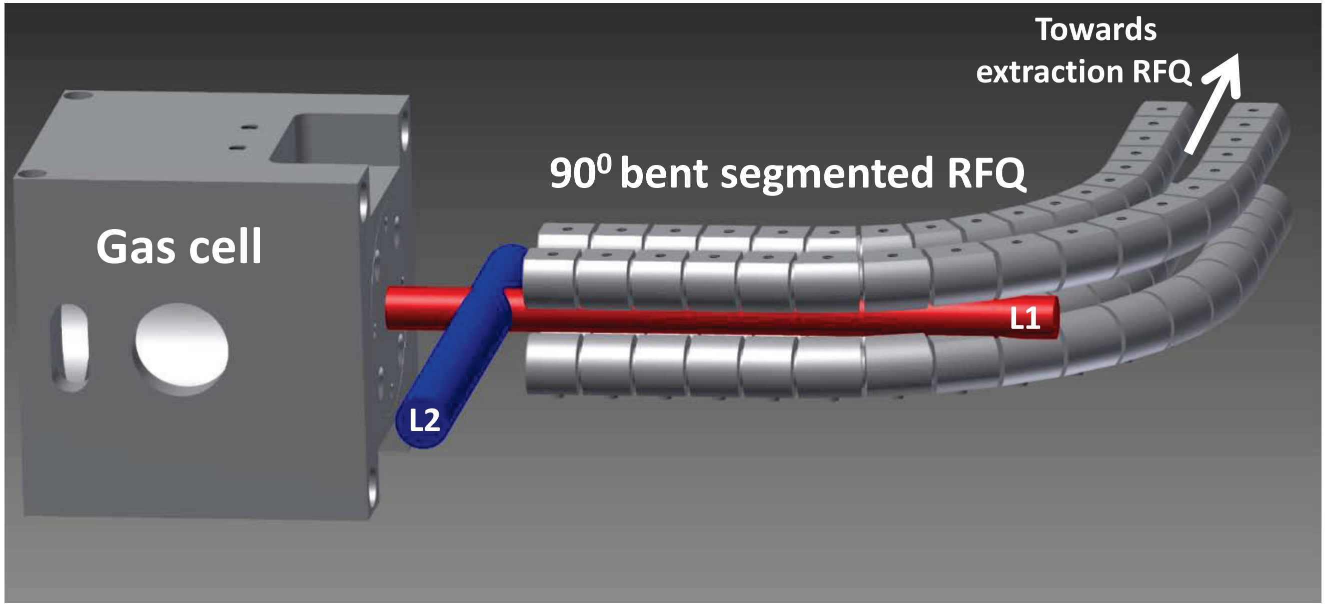


# Resonance Ionization Spectroscopy in a Free Gas Jet

Accepted NIMB, Yu. Kudryavtsev et al, http://arxiv.org/abs/1211.6649



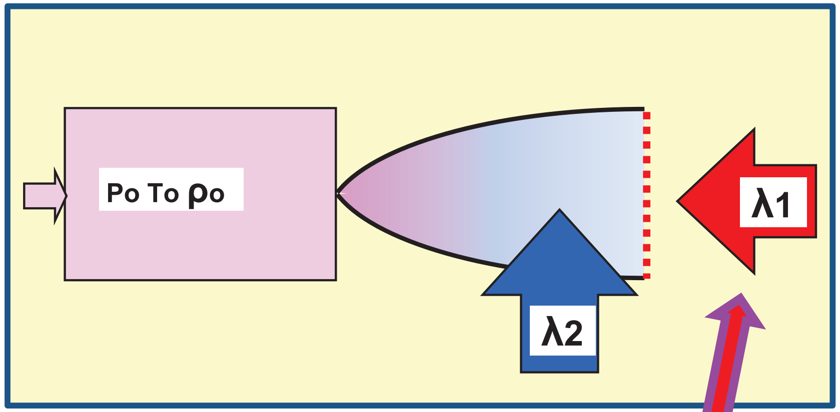


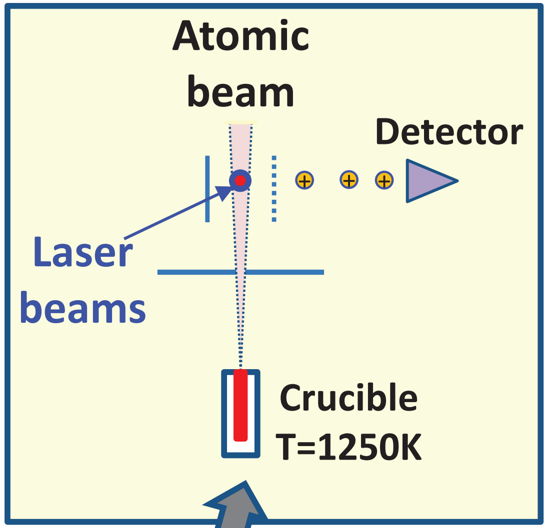


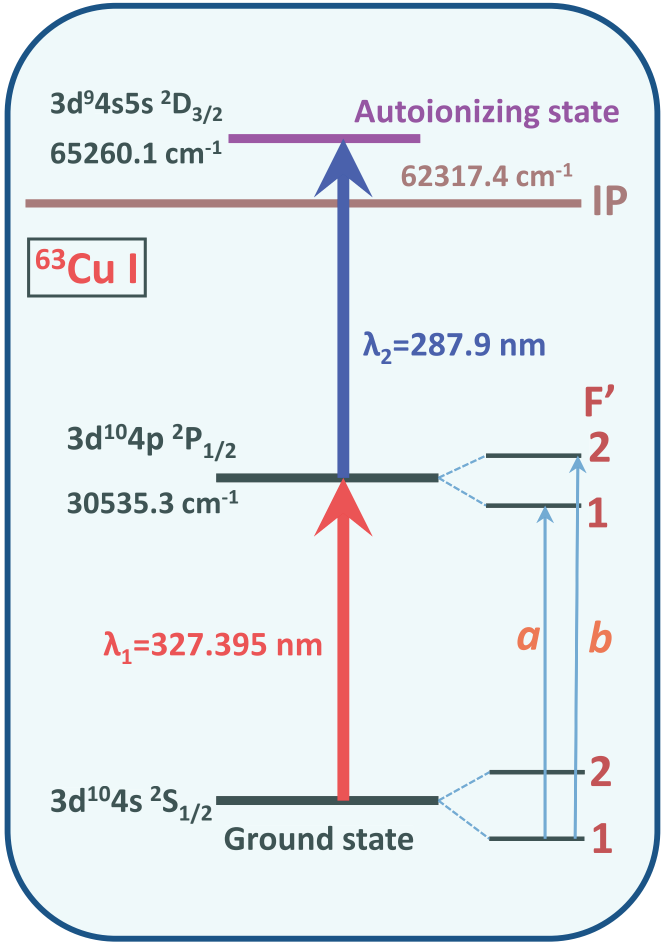
# Resonance Ionization Spectroscopy in a Free Gas Jet

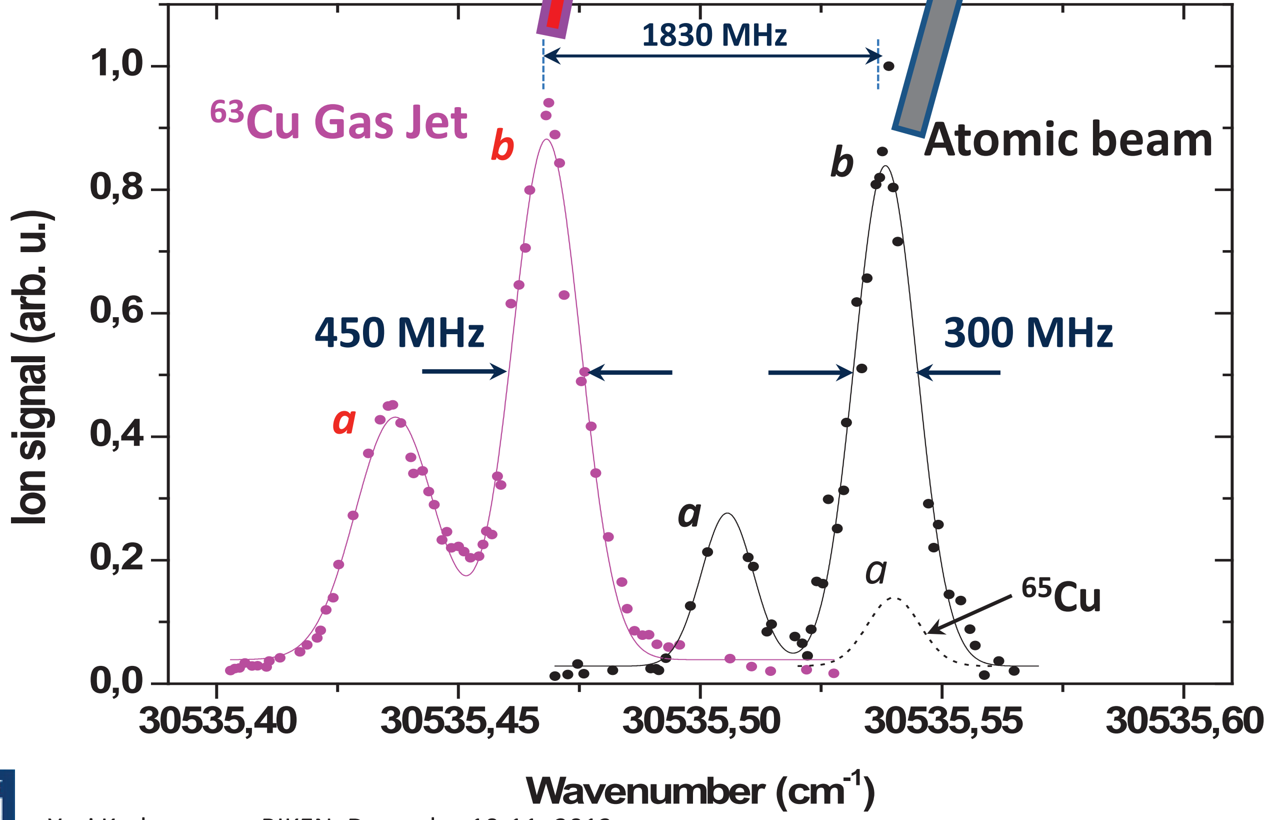
$$u = \sqrt{\frac{kT_0 \gamma M^2}{m_{ng}\left\{1+\left[\left(\gamma-1\right)/2\right]M^2\right\}}}$$

1830 MHz → $T_0 = 355 \pm 3$K









# Summary

1. **The crossed laser beams with supersonic jet has been proposed and realized at off-line conditions for two-step photo ionization in a free jet.**

2. **Using this method, the spectral resolution can be improved by more than one order of magnitude (200 MHz, Δν/ν =2.3E-7) in comparison to the gas cell.**

# Thank you for your attention