MOLECULAR ENSEMBLES IN OPTICAL LATTICES

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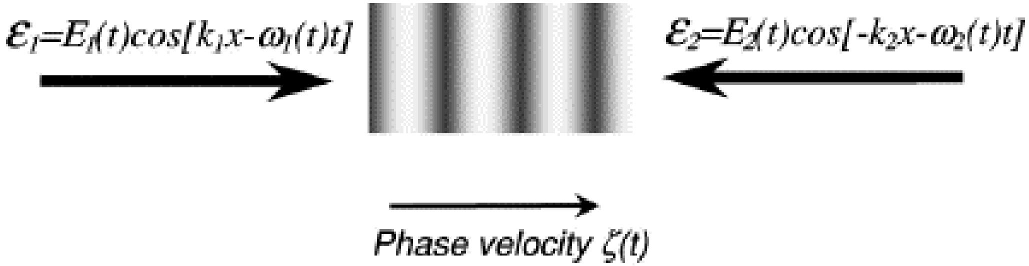
Outline

- Introduction: Optical Lattice and Gradient Dipole Force
- Coherent Raleigh-Brillouin scattering
- Microlinear acceleration and separation
- Molecular beam deceleration: theory and experiment
- Optical Landau damping and drift
- Non-resonant laser radiation absorption in the dense gas
- Conclusions

Optical Lattice



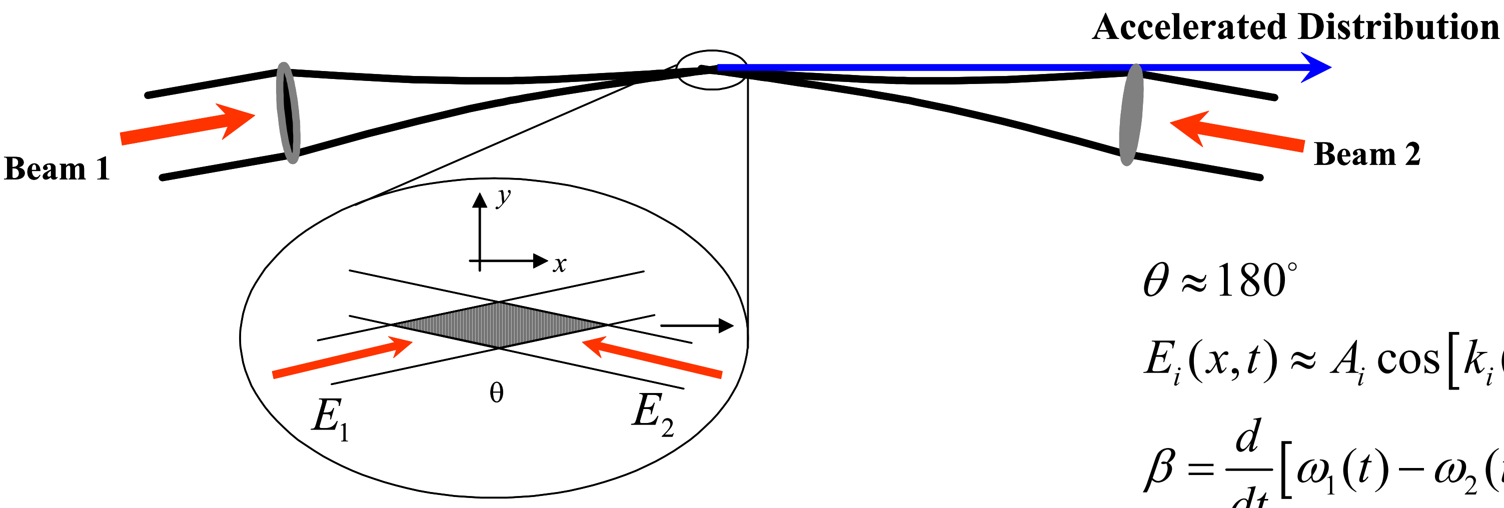
$$\Omega(t) = \omega_2 - \omega_1$$

$$\xi(t)=\Omega / q$$

$$q = \left|\vec{k}_1 - \vec{k}_2\right| \approx 4\pi / \lambda$$

Idealization: 2 laser counter propagating beams or 1 laser and a moving mirror

On practice, particles are trapped in a deep optical potential formed by two focused laser beams. To accelerate or decelerate the lattice the frequency difference between each beam is linearly chirped (β).



$$\theta \approx 180^\circ$$

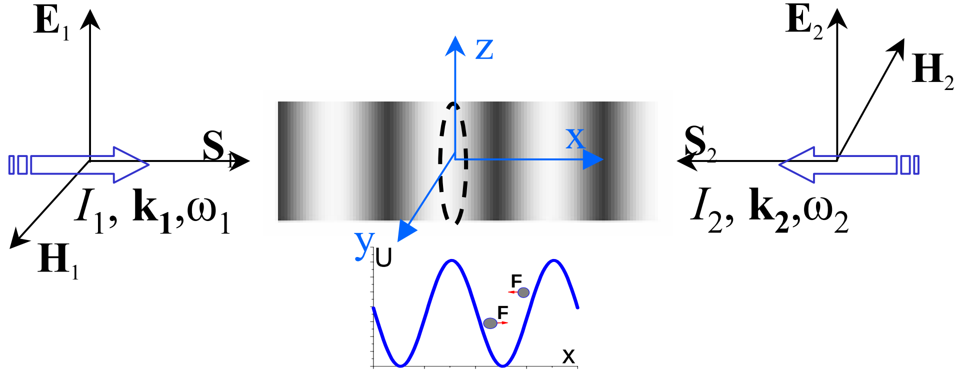
$$E_i(x,t) \approx A_i \cos\left[k_i(t)x - \omega_i(t)t\right],\ i = 1,2$$

$$\beta = \frac{d}{dt}\left[\omega_1(t) - \omega_2(t)\right]$$

$$q = \left|\vec{k}_1 - \vec{k}_2\right| \approx 2k_1$$

practical beam arrangement

Force on a particle in a lattice



- For a molecule in an optical field, the dipole moment in general case **d** =**μ** + α**E,** and the interaction Hamiltonian, H_{int} = **- d ·E**
- As high intensity pulsed fields (>10^{10} W/cm^2), deep optical potentials in the 1 to 100 K range can be created.
- For this case the Shrödinger equation reduces to the classical equation of motion, because the de Broglie wavelength of particles is much less than the spatial period of the lattice.

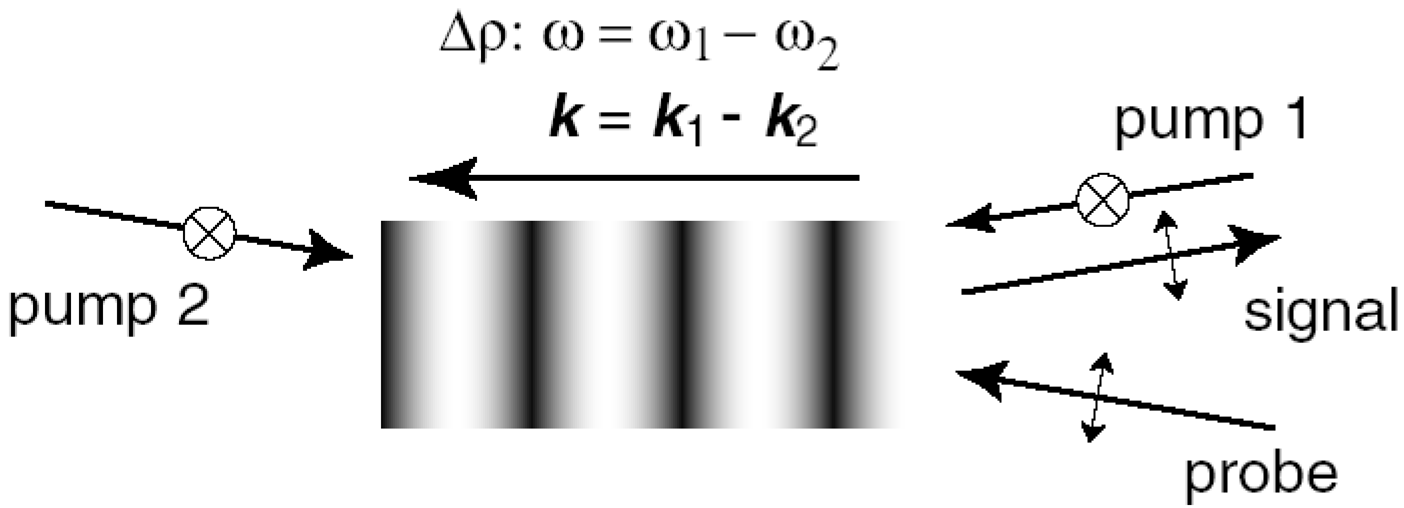
For **fields far from resonance** the dipole optical potential and force on a particle polarizability *α,* and applied field $\vec{E} = \vec{E}_1 + \vec{E}_2$, is given by:

$$U(x,t) = -\tfrac{1}{2}\alpha(\vec{E}\cdot\vec{E})$$

$$\vec{F}(x,t) = -\nabla U = -\tfrac{1}{2}\vec{i}_x \alpha q E_1(t) E_2(t)\sin[qx - \Omega(t)t]$$

Coherent Rayleigh-Brillouine scattering

Relatively low intensities of the pump beams: U<<kT; $\Delta\rho/\rho << 1$



$$I_{signal} \propto I_{pump1} I_{pump2} I_{probe} \; ,$$
$$S(\boldsymbol{k}, \omega) \propto \delta\rho^*(\boldsymbol{k}, \omega)\delta\rho(\boldsymbol{k}, \omega) \; .$$

CRBS in Monatomic Gas

- The Boltzman 1D equation in BGK approximation

$$\left[\frac{\partial}{\partial t}+v\frac{\partial}{\partial x}+\frac{F_x(x,t)}{M}\frac{\partial}{\partial v}\right]f(x,v,t)=-\frac{1}{\tau}\frac{\rho(x,t)}{\rho_0}(f-\Phi)$$

$$\Phi(x,v,t)=\frac{\rho(x,t)}{\sqrt{2\pi k_b T(x,t)}}\exp\left[-\frac{M(v-u(x,t))^2}{2k_b T(x,t)}\right]$$

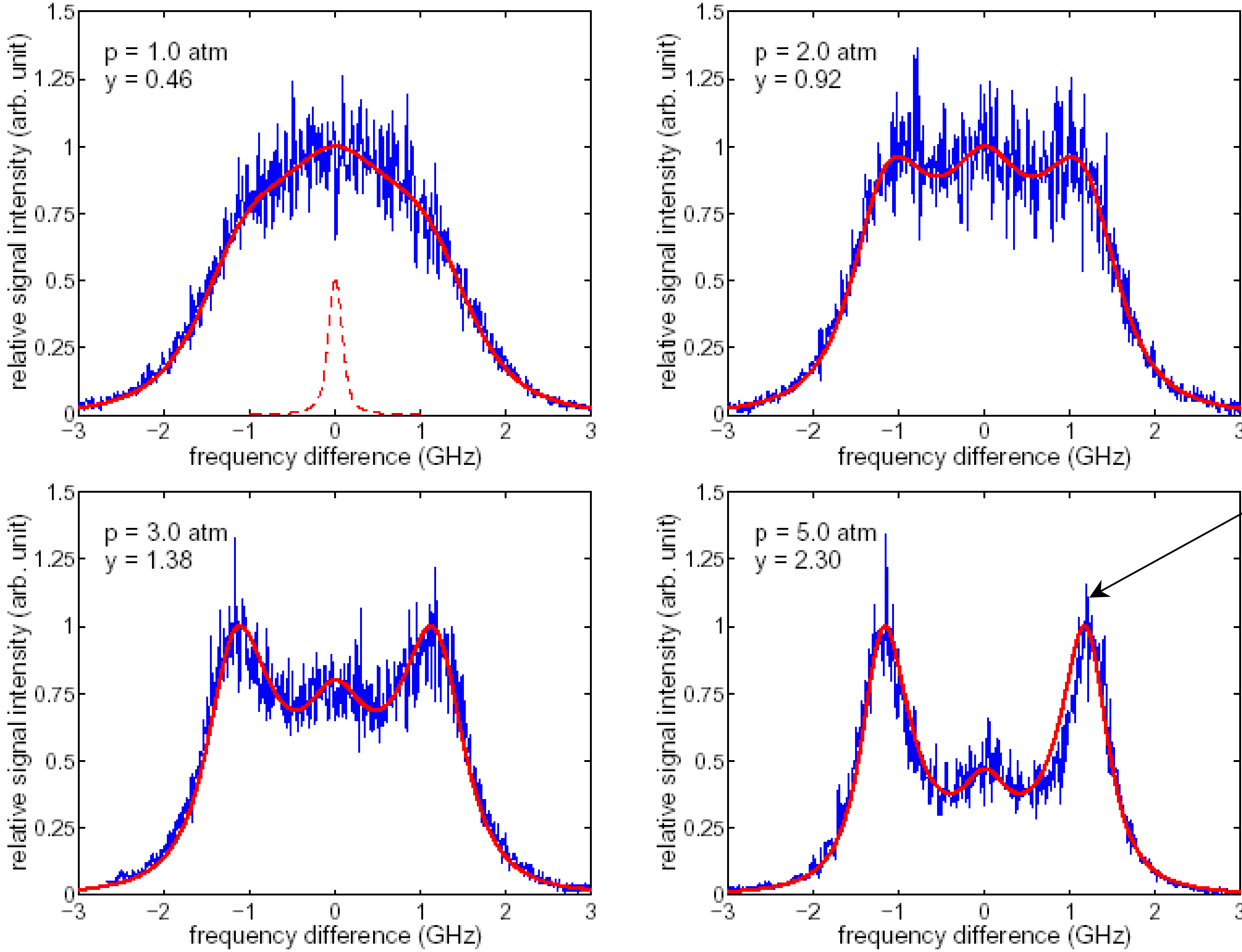
- The optical dipole force: $\vec{F}=-\nabla U;\quad U=-\frac{1}{2}\alpha E^2;\qquad E_a{}^2=2I/\varepsilon_0 c$

- BGK collision term: $f(\boldsymbol{x},\boldsymbol{v},t)\rightarrow$ *local Maxwellian distribution* Φ with relaxation time $\sim\tau$.

Coherent Rayleigh-Brillouin Scattering

Fit to argon data

T_0=292 K



$$c_s(T) = \frac{|\Delta\omega|}{q}$$

$$y = \frac{1}{q v_0 \tau} \sim \frac{\lambda}{l_n}; \quad v_0 = \sqrt{\frac{2k_B T_0}{M}}; \quad q \approx \frac{4\pi}{\lambda}$$

X.Pan, M.N.Shneider, R.B. Miles, Phys.Rev.Lett., **89**, 2002

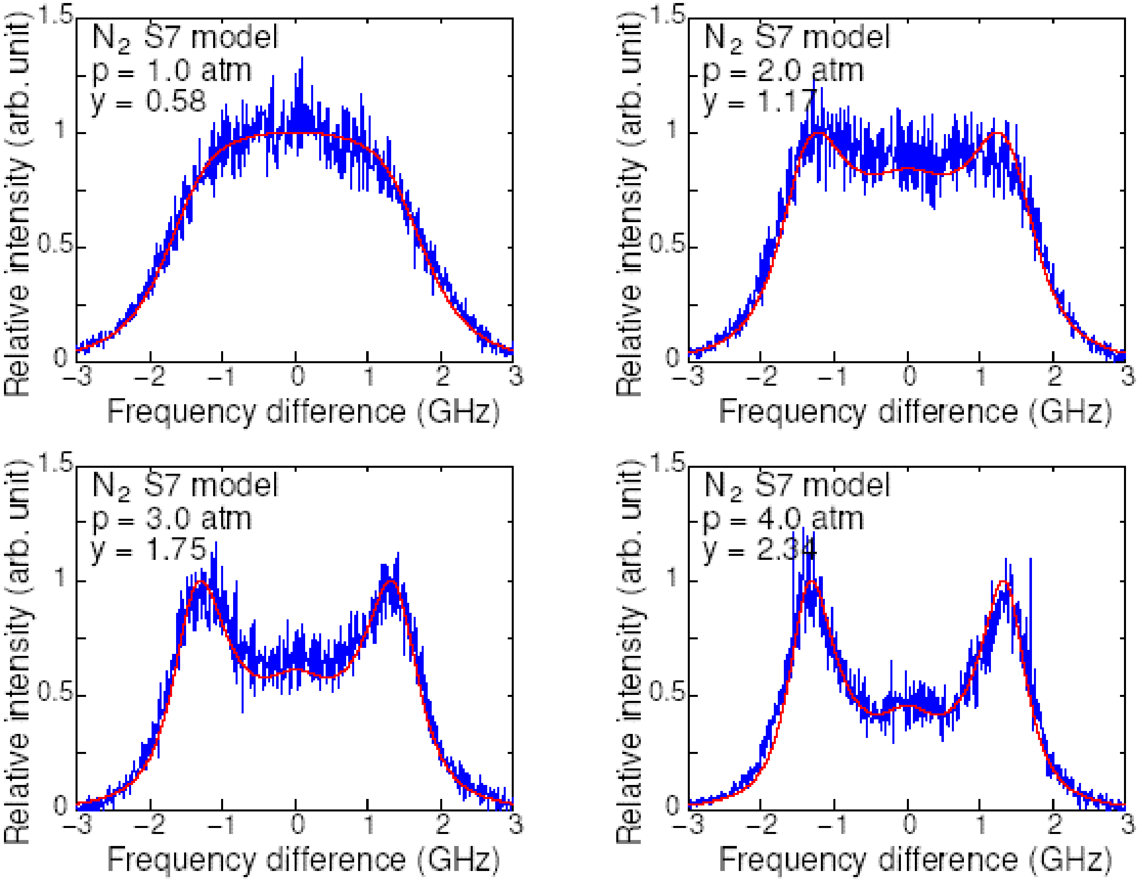
CRBS in Molecular Gas

Wang-Chang-Ulenbeck Equation

$$\frac{\partial f_i}{\partial t} + \boldsymbol{v} \cdot \nabla f_i + \boldsymbol{a} \cdot \nabla_v f_i = -\sum_{jkl} \int \int (f'_k f'_{1l} - f_i f_{1j}) |\boldsymbol{v} - \boldsymbol{v_1}| \sigma_{ij}^{kl} d\Omega dv$$

$$a = F / M \propto \alpha \nabla U \qquad i = 1, 2, \cdots$$

In equilibrium, f_i's follow the Boltzmann distribution. We solve for the density perturbation generated by the optical dipole force.



N_2 ; T_0 =292 K

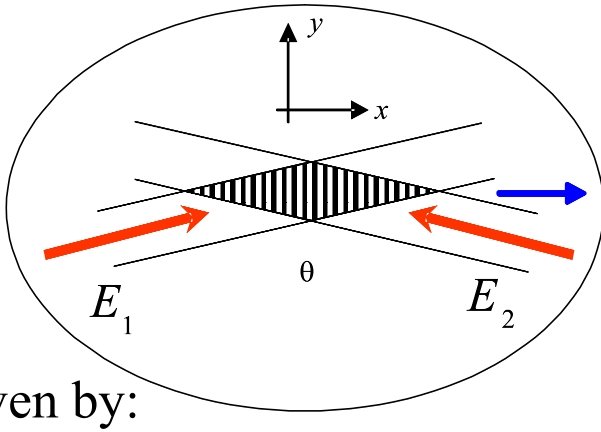
X.Pan, M.N.Shneider, R.B. Miles, Phys.Rev.A **69**, 2004

Acceleration: simplest analysis

$$U(x,t) = -\frac{1}{2}\alpha E^2, \qquad \vec{E} = \vec{E}_1 + \vec{E}_2$$

$$\vec{F}(x,t) = -\nabla U = -\frac{1}{2}\vec{i}_x \alpha q E_1(t) E_2(t) \sin[qx - \Omega(t)t]$$

$$\Omega = \omega_2(t) - \omega_1(t) = \beta t; \; \beta = d\Omega / dt; \; q = 4\pi / \lambda$$



In the lab frame the equation of motion of a particle is given by:

$$\frac{d^2 x}{dt^2} = \frac{F(x,t)}{m} = -\frac{1}{2m}\alpha q E_1(t) E_2(t) \sin(qx - \beta t^2)$$

In the accelerated frame the equation of motion is the same as that of a pendulum under constant torque and is given by:

$$\frac{d^2\theta}{dT^2} = -\frac{aq}{\beta}\sin\theta - 2, \quad \theta = qx\text{-}\Omega(t)t = qx\text{-}\beta t^2 = X\text{-}T^2$$

where θ is the phase of the particle with respect to the lattice, and $T=\beta^{1/2}t$ and $X=qx$

$$a(t) = \alpha q E(t)^2 / 2m$$

Accelerating Lattice Potential

$$\frac{d\theta}{dT} = \eta,$$

$$\frac{d\eta}{dT} = -\frac{aq}{\beta}\sin\theta - 2$$

The lattice potential is given by:

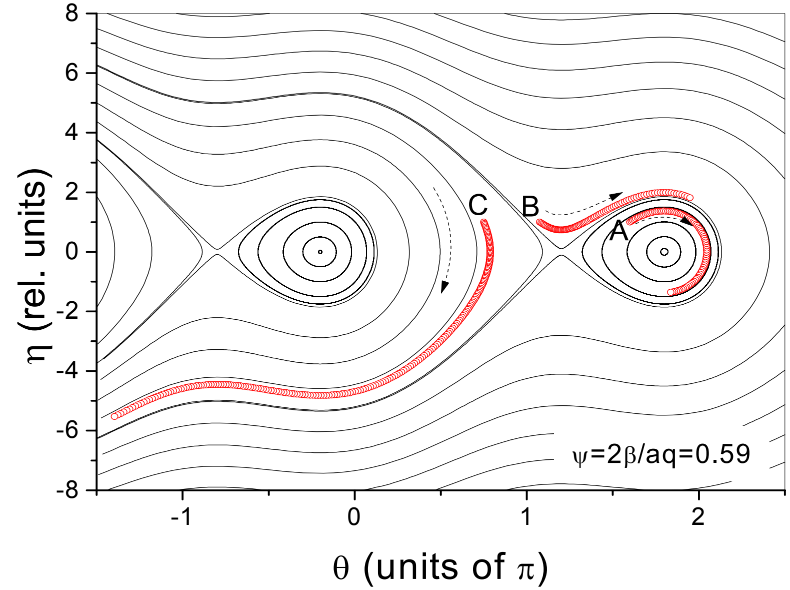
$$U(\theta) = -\int \frac{m}{q^2}\frac{d^2\theta}{dt^2}d\theta$$

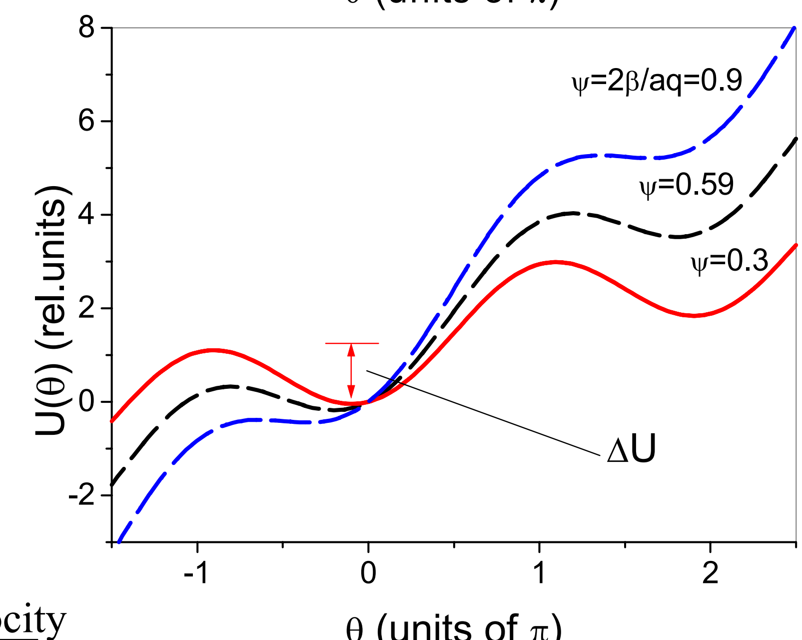
Particles can be trapped when:

$$\psi = \frac{2\beta}{aq} < 1$$

The well depth is given by:

$$\Delta U = \frac{ma}{q}\left[2\cos(\sin^{-1}\psi) - \psi(\pi - 2\sin^{-1}\psi)\right]$$





A particle can be trapped and accelerated if its velocity differs from the lattice velocity by up to : $\Delta v = \sqrt{\frac{2\Delta U}{m}}$

P.F.Barker, M.N.Shneider, Phys.Rev A **64**, 2001

Acceleration of an ensemble (top hat temporal profile)

$$\frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x} + \frac{F(x,t)}{m}\frac{\partial f}{\partial v} = \left(\frac{\partial f}{\partial t}\right)_c \qquad \left(\frac{\partial f}{\partial t}\right)_c = 0$$

CH_4; p=0.1 Torr; T_0=300 K.

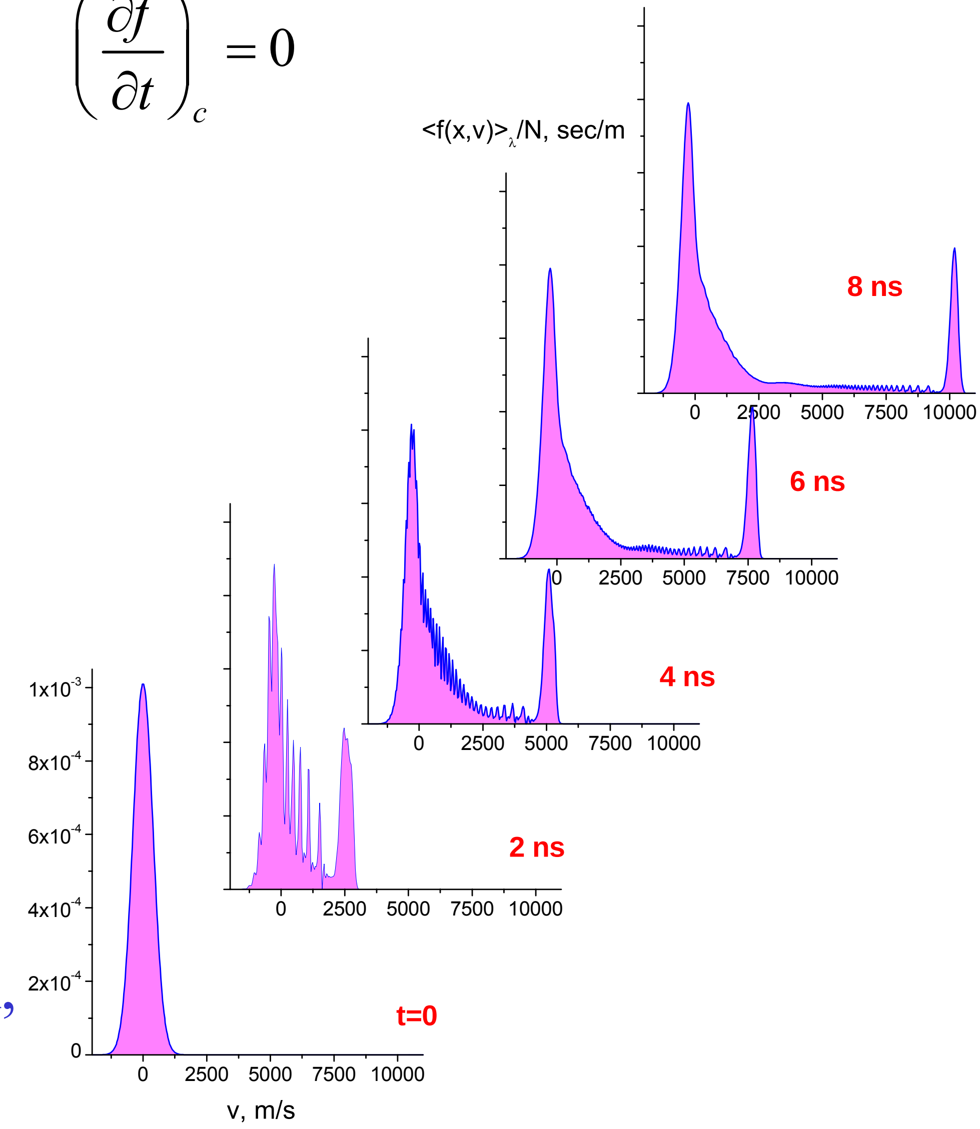
Approximately 30 % of the molecules are accelerated to a velocity of 10.1 km/s

For this calculation :

$\psi = 0.59$, $q = 1.57\text{x}10^7$ m^{-1},

$a = 2.14\text{x}10^{12}$ m/s^2, $\Delta U = 133$ K,

$I = 3\text{x}10^{12}$ W/cm^2.

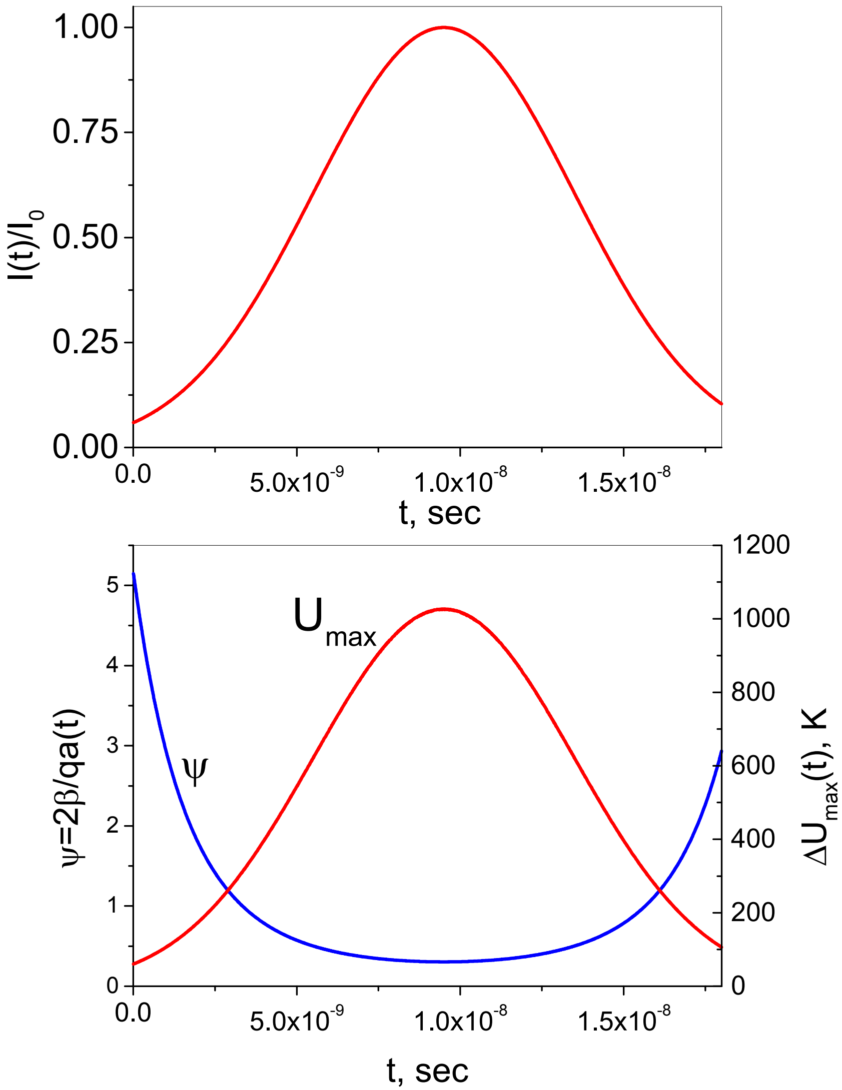


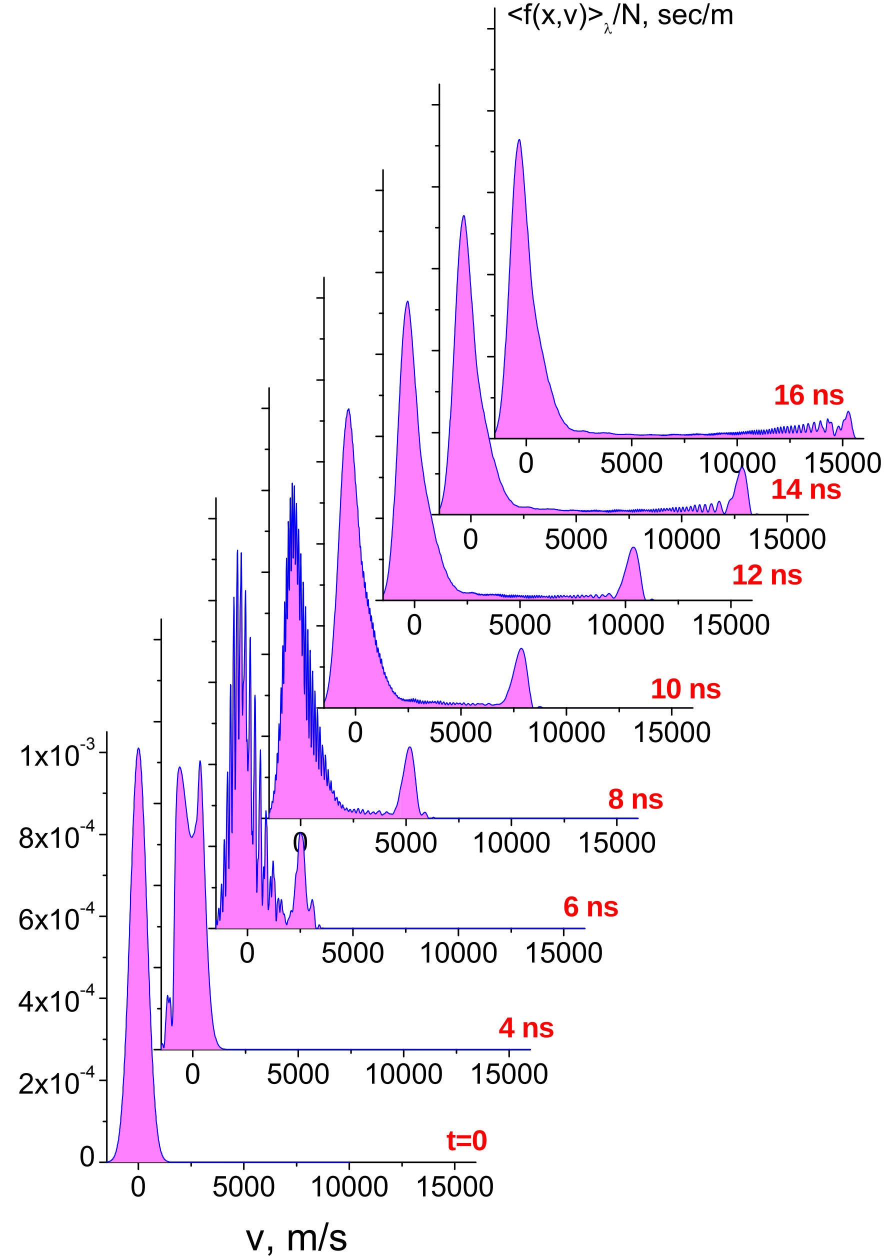
P.F.Barker, M.N.Shneider, Phys.Rev A **64**, 2001

Acceleration of CH_4 (Gaussian temporal profile)

CH_4; p=0.1 Torr; T_0=300 K.

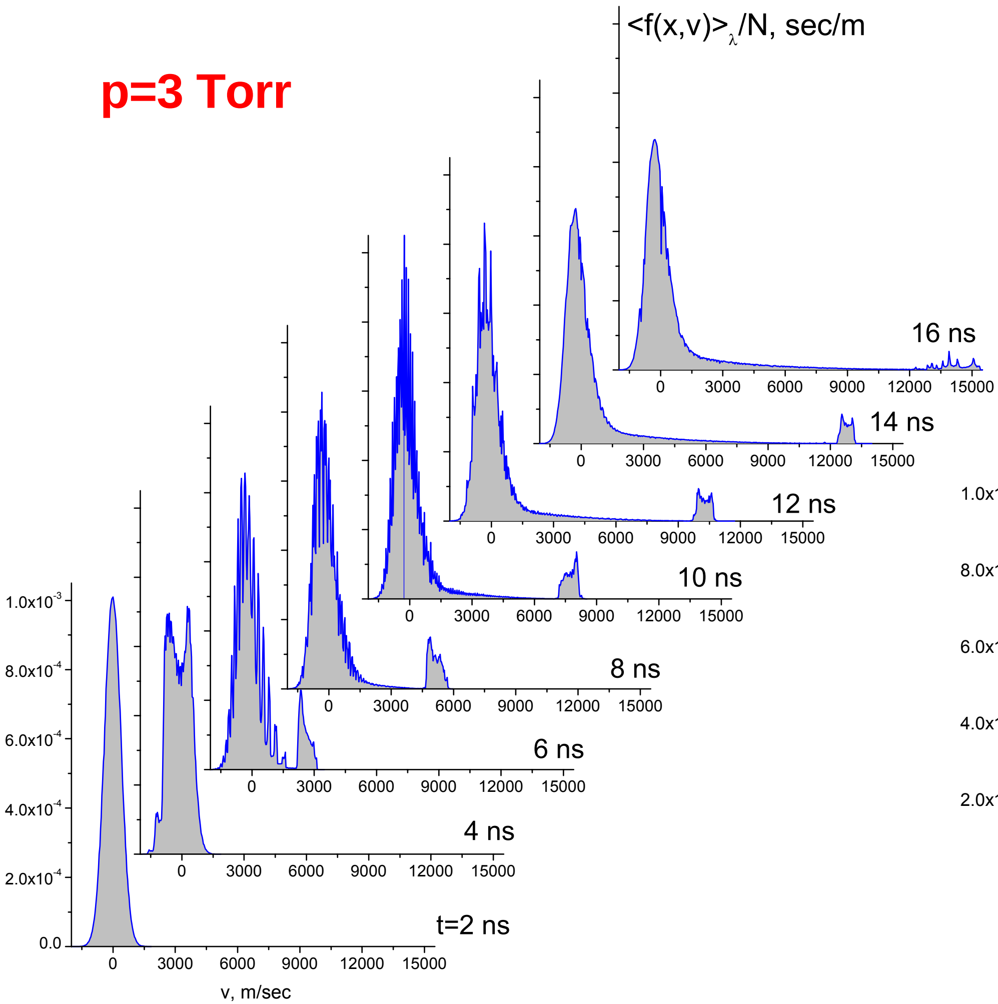
ξ_0 = - 5 km/s.

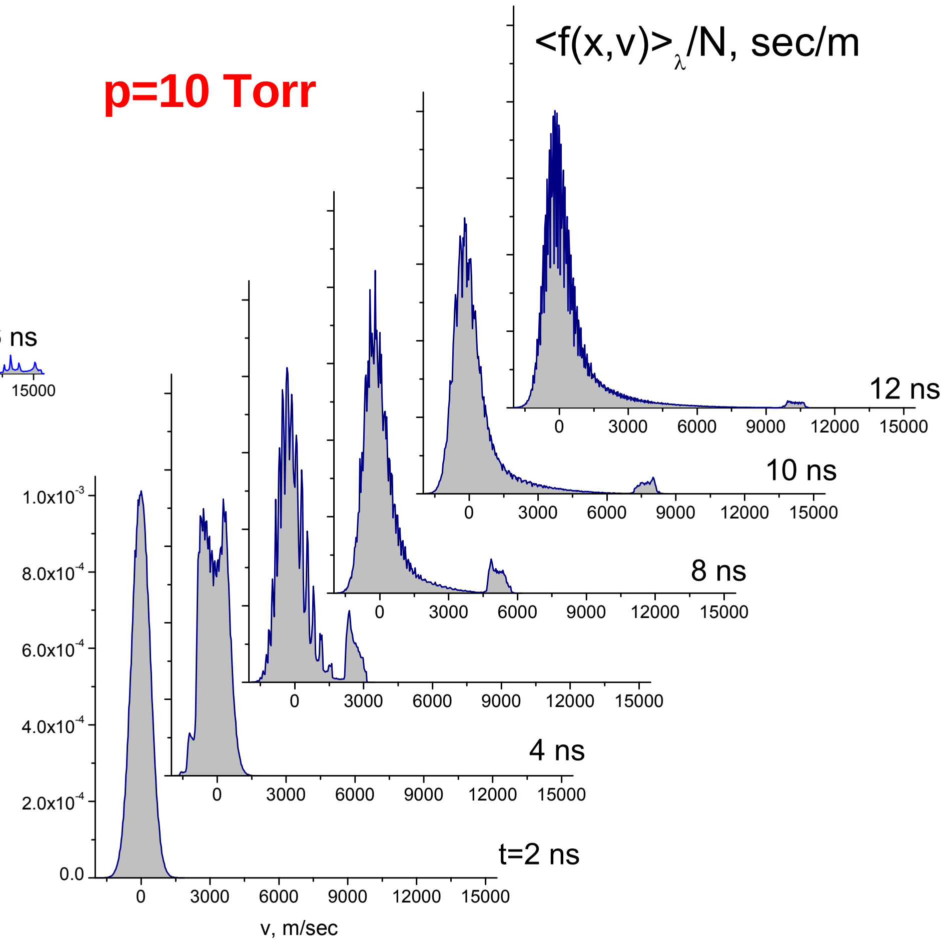




P.F.Barker, M.N.Shneider, Phys.Rev A **64**, 2001

Collisional acceleration of CH_4 (Gaussian)
DSMC calculations: CH_4, T_0 =300 K

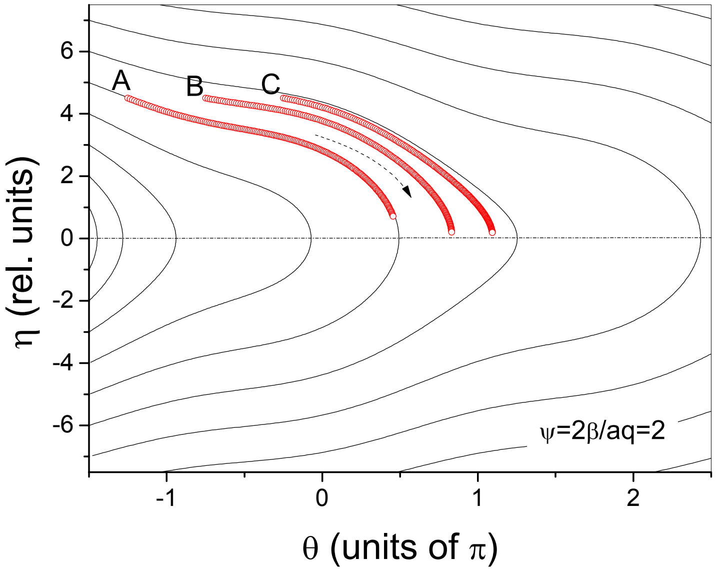




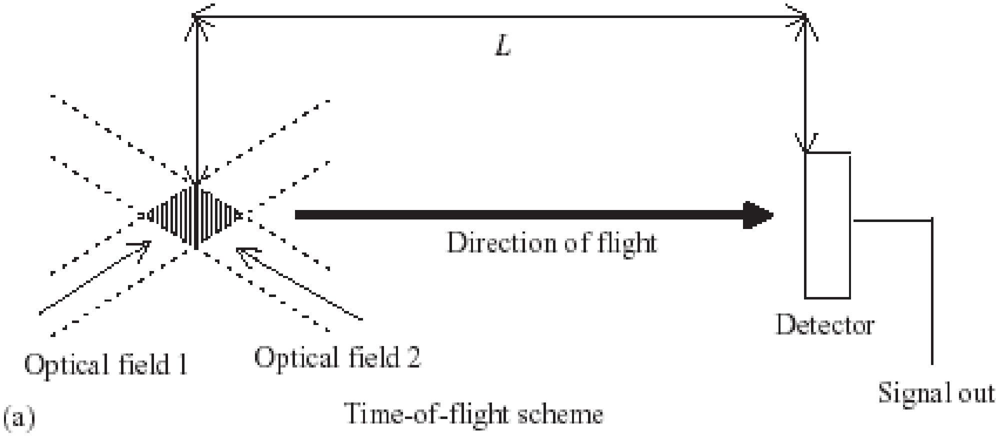
Performed in collaboration with Sergey Gimelshein (USC)

Untrapped particle acceleration: $\psi > 1$

An ensemble of particles can be strongly perturbed by the lattice even when no potential well exists ($\psi > 1$)



P.F.Barker, M.N.Shneider, Phys.Rev A 64, 2001

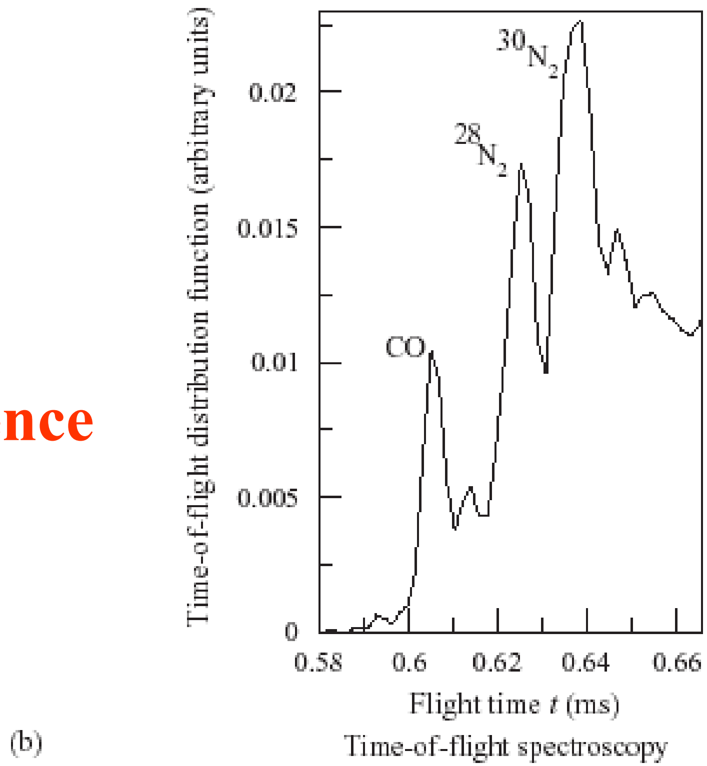


Separation based on α/M difference

Example:

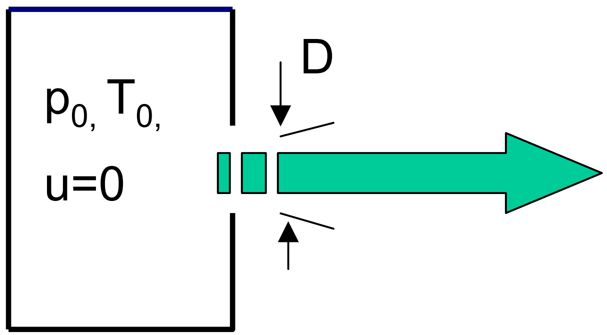
Molecular beam: $^{30}N_2 : ^{28}N_2 : CO = 1:1:1$; $v_b = 239.1$ m/s; T=5 K

Optical lattice: $I = 3.3 \times 10^{11}$ W/cm^2; $\beta = 5.47 \times 10^{17}$ rad/sec^2 ; $\Delta t = 5$ ns



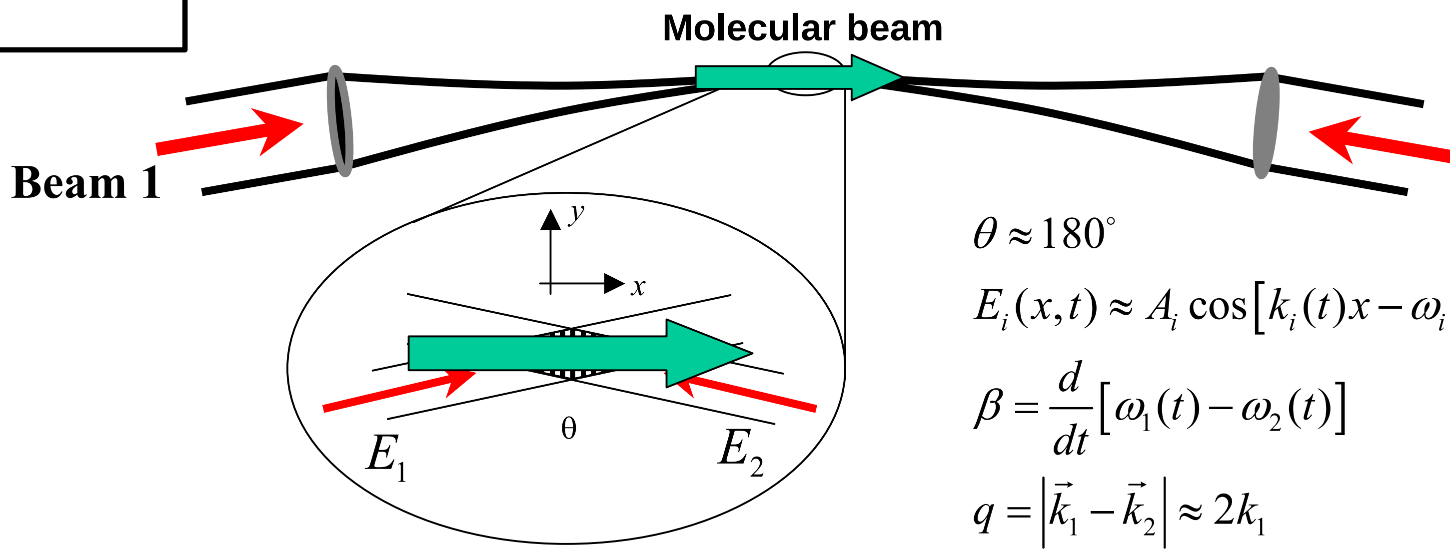
G. Dong, W. Lu, P. F. Barker, M.N.Shneider, Progress Quant. Electronics, **29** 2005

Molecular beam trapping and slowing down



$$H(x)=0.5u^2 + c_pT(x) = c_pT_0$$

$$\text{At } x/D>5;\ \ v_b \sim u_\infty = c_0[2/(\gamma-1)]^{1/2} = [2(\gamma RT_0)/(\gamma-1)]^{1/2}$$



$$\theta \approx 180^\circ$$

$$E_i(x,t) \approx A_i \cos\left[k_i(t)x - \omega_i(t)t\right],\ i = 1,2$$

$$\beta = \frac{d}{dt}\left[\omega_1(t) - \omega_2(t)\right]$$

$$q = \left|\vec{k}_1 - \vec{k}_2\right| \approx 2k_1$$

The OL phase velocity $\xi = \Omega/q$ reduces from $\xi = v_b$ to $\xi = 0$ during the pulse.

This scheme can be used for deceleration with the potential to bring to rest supersonically cooled molecules with temperature < 1 K. A high density of stationary cold molecules could be created with densities in the 10^{13} - 10^{15} cm^{-3} range.

Suggested by Barker and Shneider (Phys.Rev A **65**, 2002)

The deceleration of an ensemble of particles

Boltzmann equation:

$$\frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x} + \frac{F(x,t)}{m}\frac{\partial f}{\partial v} = \left(\frac{\partial f}{\partial t}\right)_c = 0$$

$$f(t=0,v) = \frac{\exp[-M(v-v_b)^2/2k_bT_0]}{\sqrt{2\pi k_b T_0/m}}$$

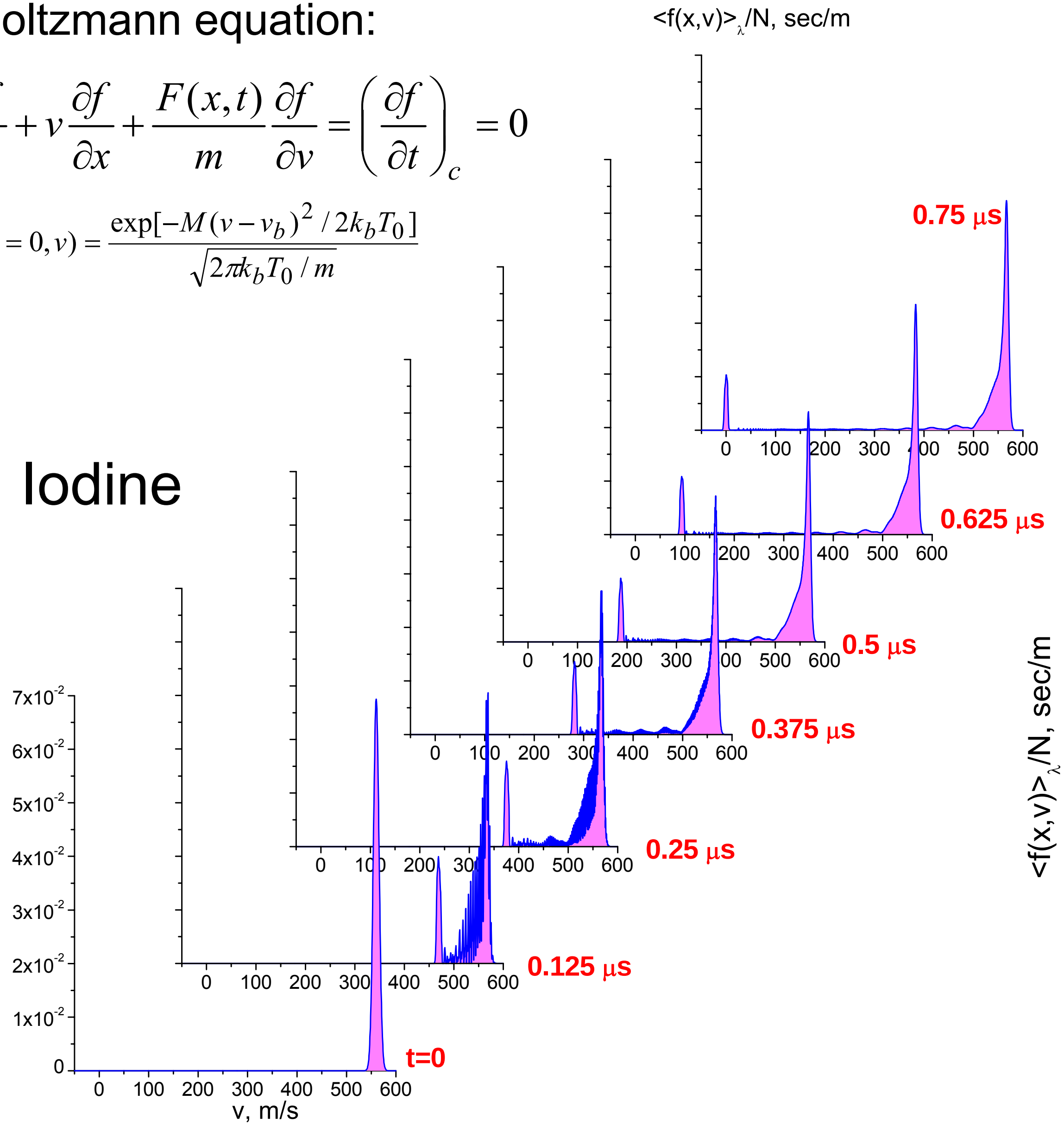
$I=1.2\times10^{14}$ W/m^2; $\psi=0.735$

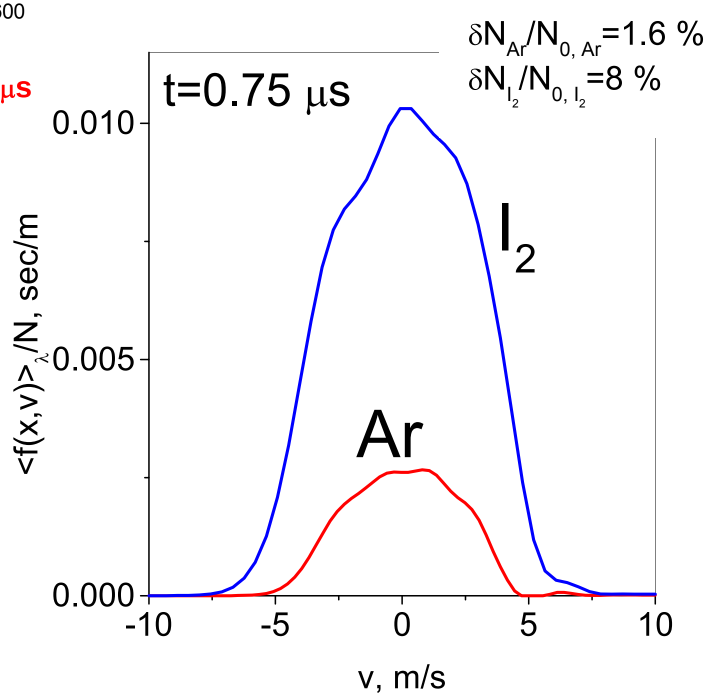
$v_0=560$ m/s; $T_0=1$ K

I_2 : $U_{max} = \alpha I z_0 = 1$ K

Ar: $U_{max}=0.157$ K

Iodine



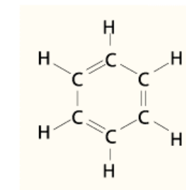


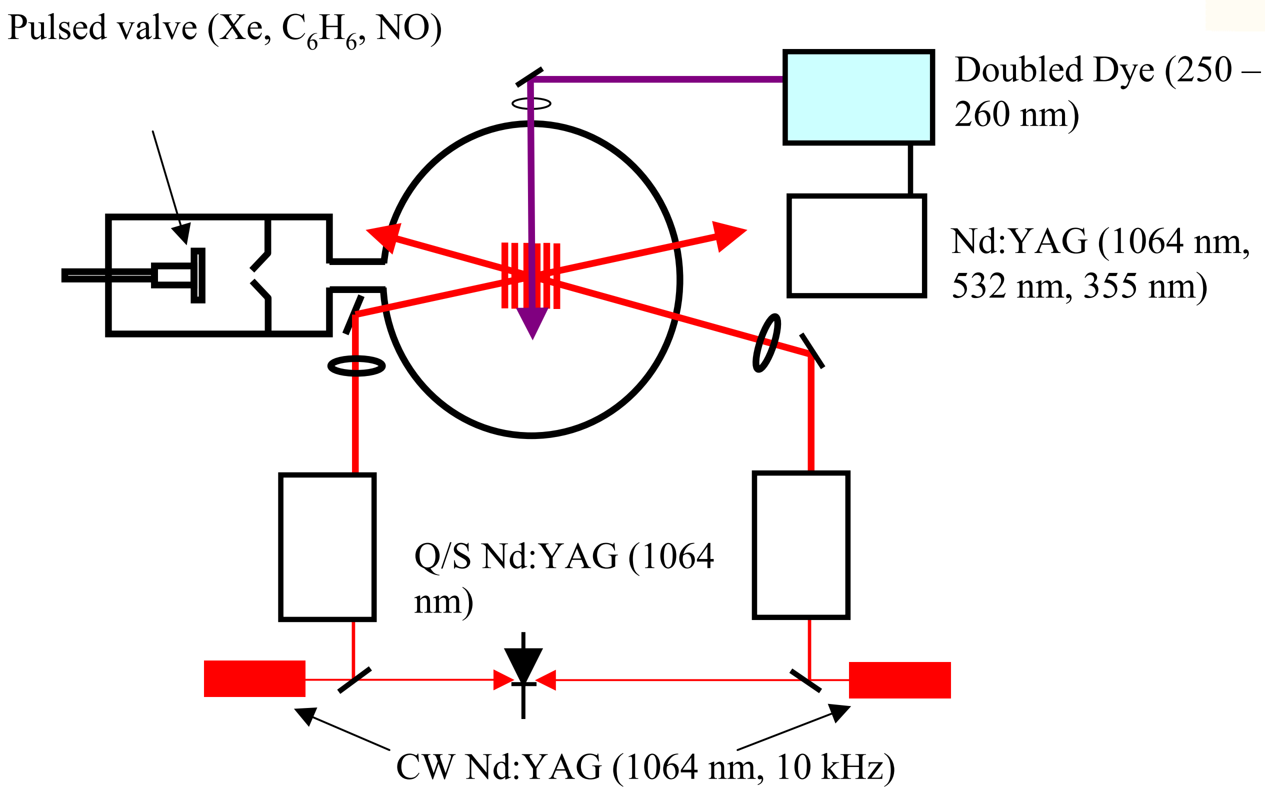
P.F.Barker, M.N.Shneider, Phys.Rev A 65, 2002

Newest experimental results: Heriott-Watt University 2004

Peter Barker, Ray Fulton and Alex Bishop

Optical Lattice affected on benzene (C_6H_6) molecular beam

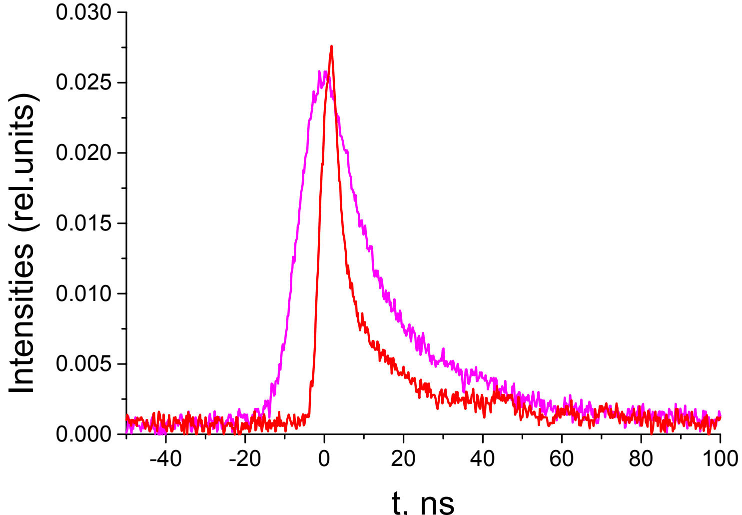




- $I_{IR} \sim 5\text{x}10^{11}$ Wcm^{-2} ; 1/e^2 radius = 60 μm
- $I_{UV} \sim 10^7$ Wcm^{-2} ; 1/e^2 radius = 4 μm

R.Fulton, A.Bishop, M.N.Shneider, P.F.Barker (submitted to PRL,2005)

Pulsed optical lattice



Measured laser beam intensities.
Energy per pulse: $\Sigma_1 \approx \Sigma_2 \approx 0.345$ J

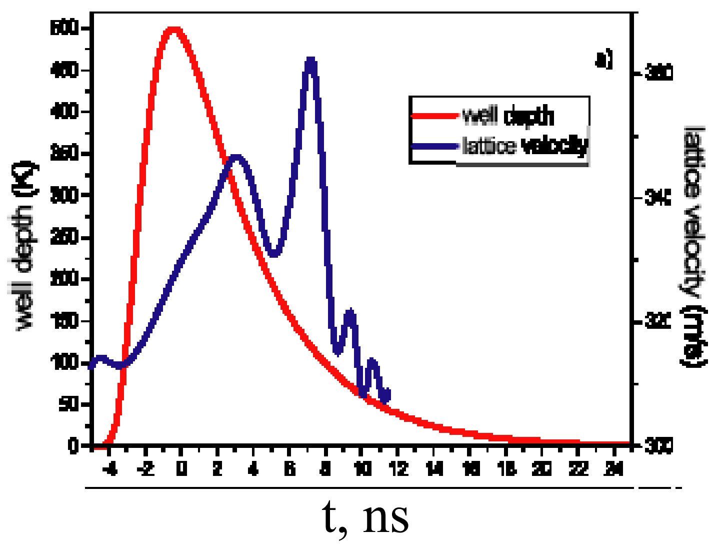
$$\frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x} + \frac{F(x,t)}{m}\frac{\partial f}{\partial v} = \left(\frac{\partial f}{\partial t}\right)_c = 0$$

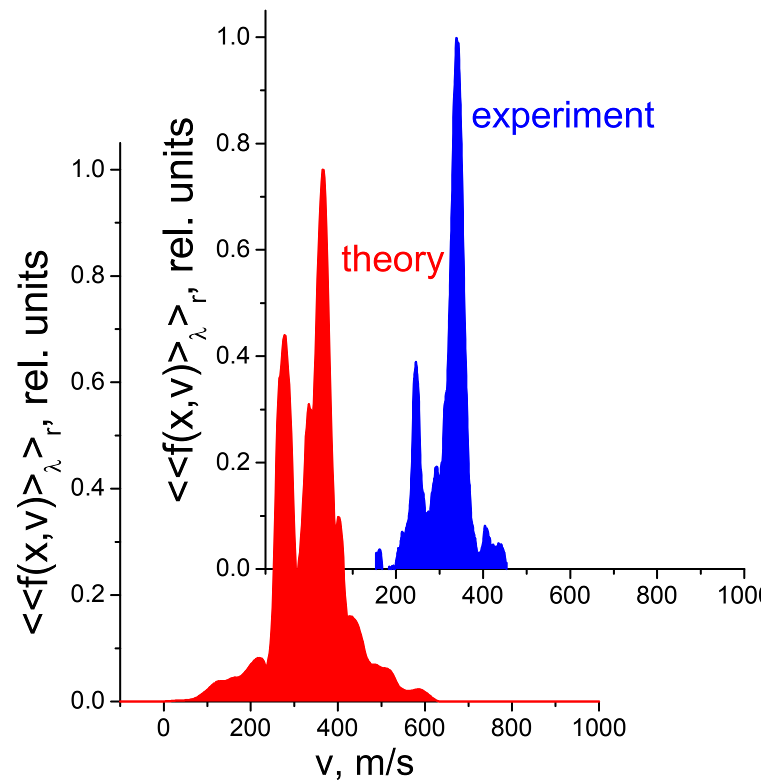
$$F = -\nabla U$$

$$U = -\frac{\alpha}{\varepsilon_0 c}\sqrt{I_1(r,t) I_2(r,t)}\cos(kx - \Delta\omega t)$$

$$f(t=0,v) = \frac{\exp[-M(v-v_b)^2/2k_b T_0]}{\sqrt{2\pi k_b T_0/m}}$$

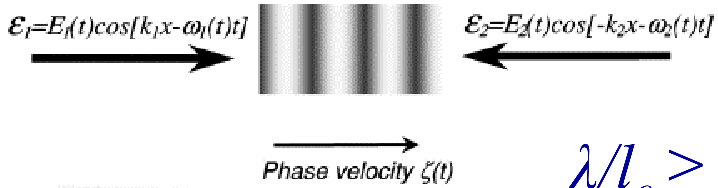
$$T_0 = 2.3\ \text{K};\quad \text{v}_\text{b} = 320\ \text{m/s}$$





R.Fulton, A.Bishop, M.N.Shneider, P.F.Barker (submitted to PRL,2005)

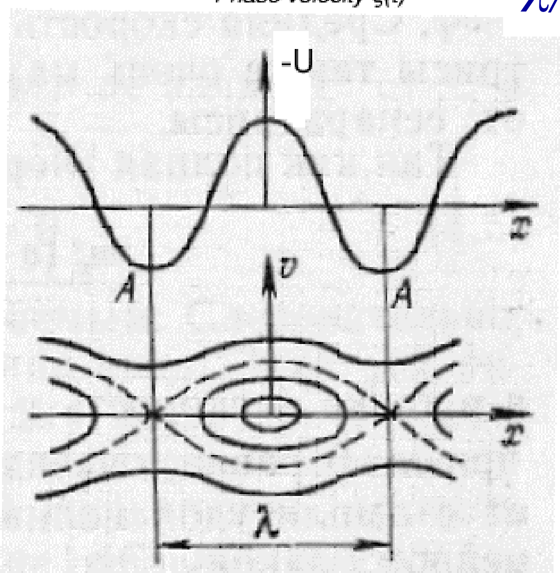
Optical drift and Landau damping

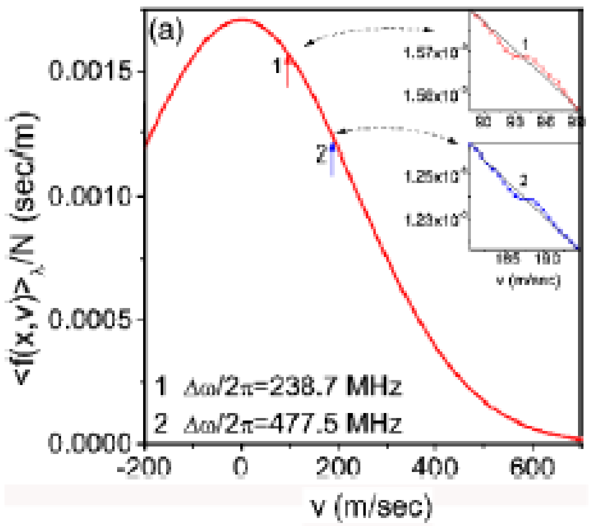


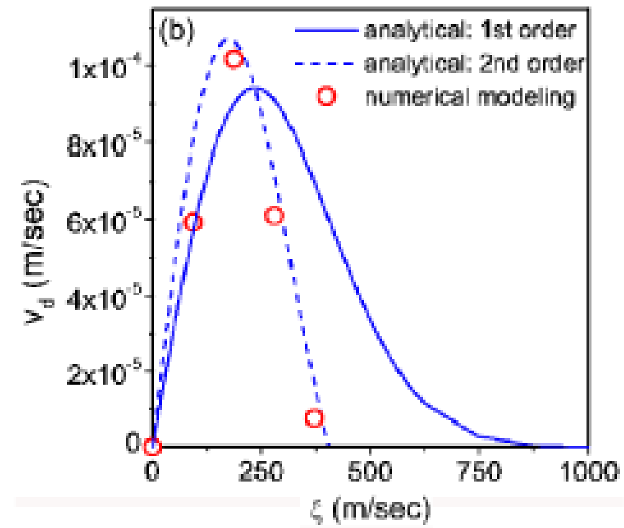
$\lambda/l_c \geq 1$

Rb; p=100 Pa; T=560 K; W=100 mW; D=40 μm

(I=7.95×10^7 W/m^2; U=50 mK)







$$\Omega = \omega_2 - \omega_1 = \text{const};\ \xi = \Omega/q = \text{cons}$$

$$F \propto \alpha q E_1 E_2 \sin(qx - \Omega t)$$

$$\frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x} + \frac{F(x,t)}{m}\frac{\partial f}{\partial v} = -\frac{f - f_0}{\tau}$$

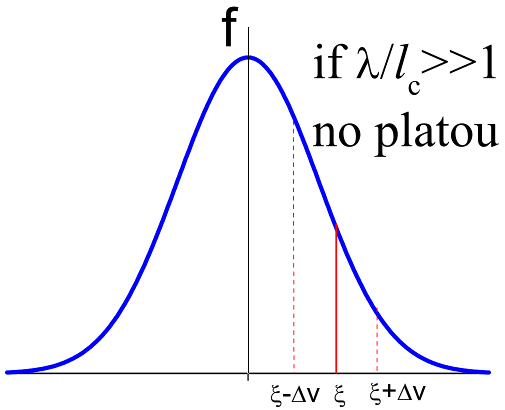
$$\text{Drift velocity}: \mathrm{v}_{\mathrm{dr}} = \left\langle \frac{1}{N}\int_{-\infty}^{\infty} f(x,v,t)\,v\,dv \right\rangle_\lambda$$

Optical Landau damping, where atoms with velocities less than the phase velocity take energy from the field, and faster atoms give energy to the field through stimulated scattering. **In result: plateau formation.**

Plateau at $\xi-\Delta v < v < \xi+\Delta v$; $\Delta v = (2U/m)^{1/2}$

Shneider, Barker, Phys.Rev. A, **71,** 2005

Non-resonant laser radiation absorption in the dense gas



Relaxation time in the gas: $\tau \approx \tau_{col.}$ In Air at p ~ 1 Atm, T~300 K, l_c < 100 nm

The energy density of OL electromagnetic wave: $W = \frac{\varepsilon_0 E_a^2}{2} = \frac{I}{c}$

The change of kinetic energy in collision with wall: $\Delta\varpi = 2M(v-\xi)\xi$

The power, transferred from the particle to the wall:

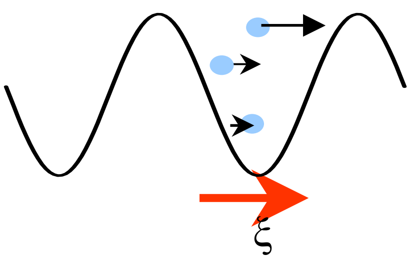
$$\Delta\dot{\varpi} = \Delta\varpi/\tau_{col} = 2M(v-\xi)^2\xi/l_c$$

The total rate of the gas-OL energy exchange: $\frac{dW}{dt} \approx -\int_{\xi-\Delta}^{\xi+\Delta} \Delta\dot{\varpi}(v) f(v)dv$

$$f(v) \approx f_0(\xi) + \frac{df_0}{dv}_{\xi}(v-\xi)$$

$$\Delta = \sqrt{2\phi_m/M}\ ,\ \phi_m = \alpha I z_0$$

The maximal dissipation rate at: $\xi_{max} = \sqrt{2k_B T/M}$



Potential well mowing with phase velocity, ξ

The rate of the momentum transfer per particle:
$d\Delta p/dt \approx \Delta p/\tau_{col} = 2M(\xi - v)|\xi - v|/l$

Rate of the momentum transfer:

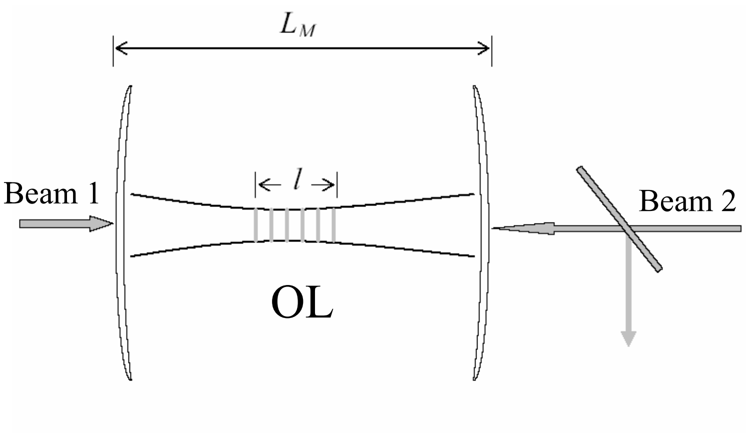
If gas is moving along the OL axis with the velocity v_z:

$$P_d = \frac{m^2(\xi-v_z)^2}{l_c k_B T} f_0(\xi - v_z)\Delta^4;$$

$$F_z = \frac{m^2(\xi-v_z)}{l_c k_B T} f_0(\xi - v_z)\Delta^4;$$

Shneider, Barker, Phys.Rev. A, **71,** 2005

Non-resonant laser radiation absorption in the dense gas



Experimental arrangement to measure optical Landau damping within a near concentric optical cavity

The temporal variation in intensity of a pulsed field in the cavity: $dI/dt = -I/\tau_Q - N\sigma_R Ic - 2P_d cl/L_M$

The empty cavity lifetime: $\tau_Q = \frac{L_M}{c(1-R)}$

The Raleigh scattering losses: $N\sigma_R Ic, \quad \sigma_R = \frac{8\pi^3\alpha^2}{3\varepsilon_0^2\lambda^4}$

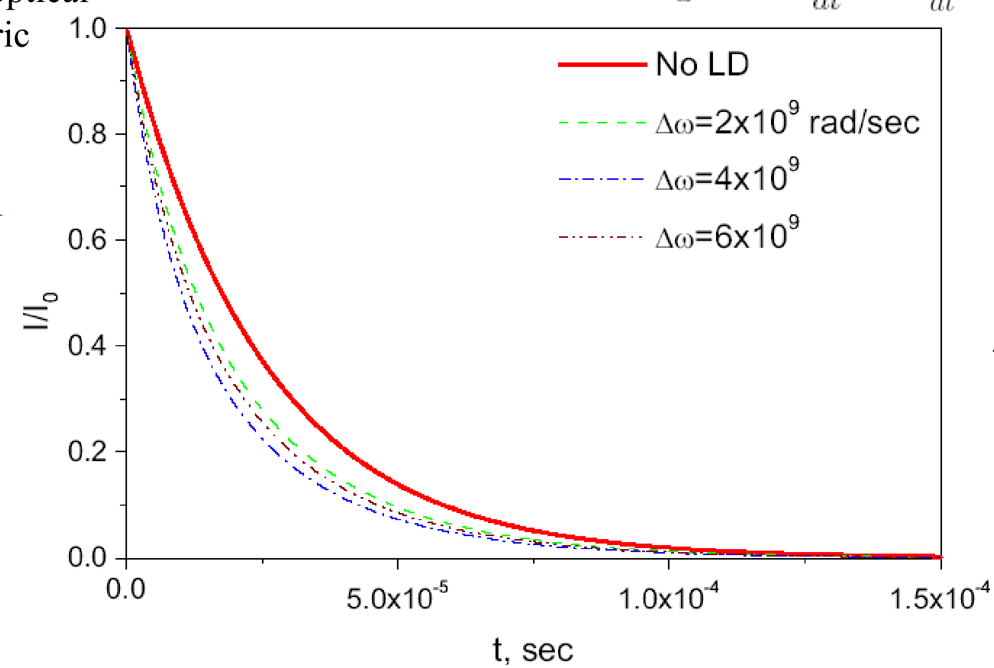
Landau dissipation rate: $P_d = -\frac{dW}{dt}; \quad \frac{dW}{dt} = -\frac{M^2\xi^2}{l_c k_B T} f_0(\xi)\Delta^4$

Ar: T_0=293 K; p=50 Atm

$I_0 = 5\text{x}10^{12}$ W/m^2

R=0.99995; λ=1.064 μm

L_m=50 cm; l=2.5 cm

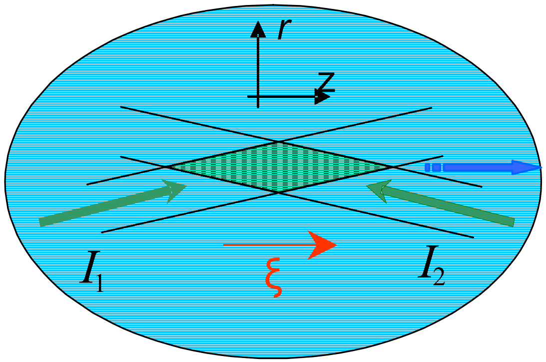


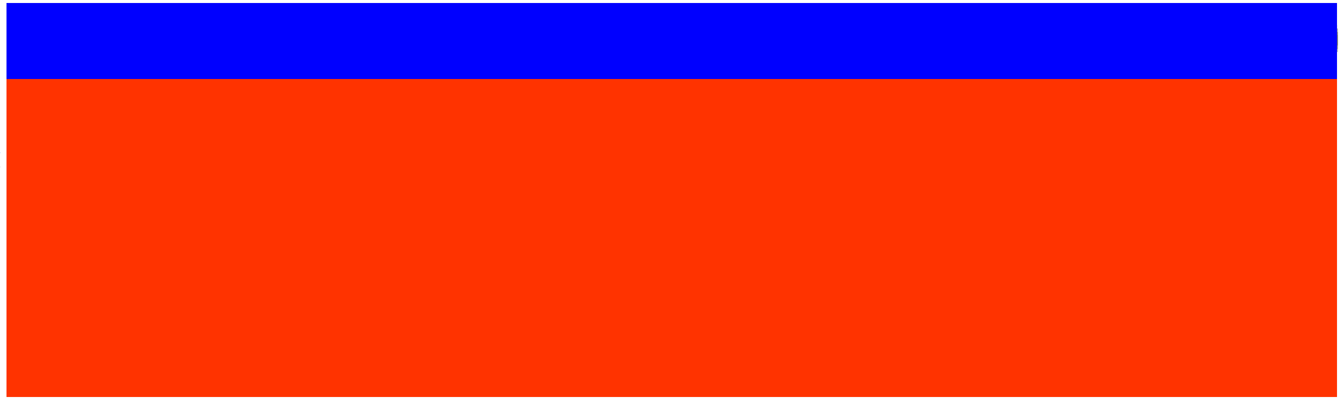
$\Delta\omega \approx 4\pi\xi/\lambda$

Laser intensity as a function of time in the cavity with and without Landau damping

Shneider, Barker, Phys.Rev. A, **71,** 2005

Formation of localized gas jets in “free space”





The full set of Euler gasdynamic equations in cylindrical coordinates

$$\frac{\partial}{\partial t}\left\|\begin{matrix}\rho\\ \rho v_r\\ \rho v_z\\ e\end{matrix}\right\| + \frac{\partial}{\partial r}\left\|\begin{matrix}\rho v_r\\ \rho v_r^2+p\\ \rho v_z v_r\\ (e+p)v_r\end{matrix}\right\| + \frac{\partial}{\partial z}\left\|\begin{matrix}\rho v_z\\ \rho v_r v_z\\ \rho v_z^2+p\\ (e+p)v_z\end{matrix}\right\| = -\frac{1}{r}\left\|\begin{matrix}\rho v_r\\ \rho v_r^2\\ \rho v_z v_r\\ (e+p)v_r\end{matrix}\right\| + \left\|\begin{matrix}0\\ F_r\\ F_z\\ P_d\end{matrix}\right\|$$

$$e = \rho\left[\varepsilon + (v_r^2 + v_z^2)/2\right], \qquad p = (\gamma - 1)\rho\varepsilon$$

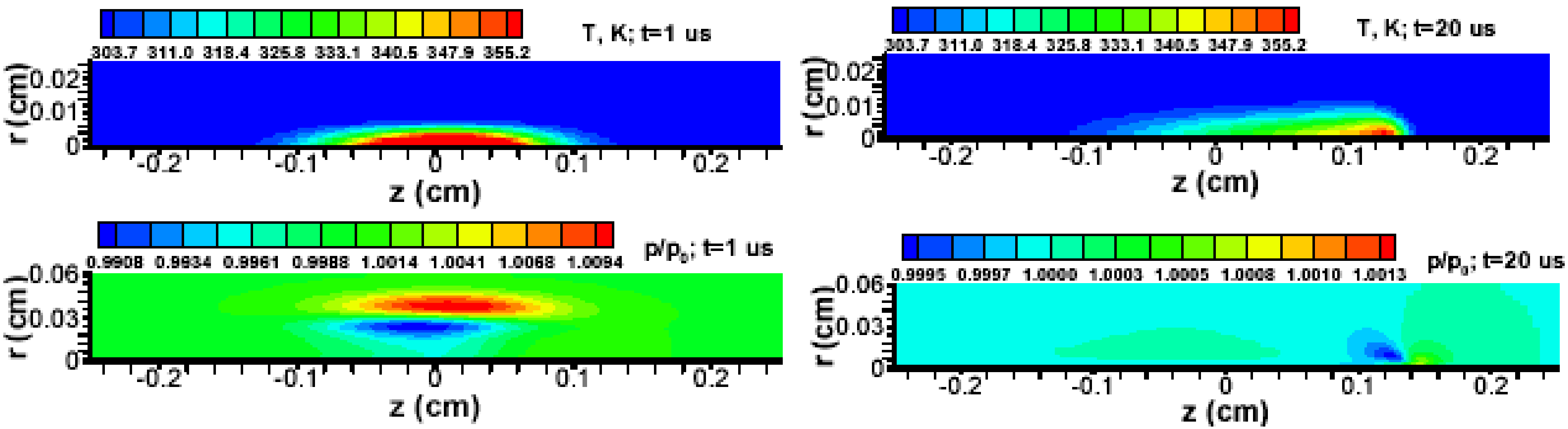
$$F_z(r,z,t) = \dot{\Theta}, \text{ and } F_r(r,z,t) = -\frac{\partial U}{\partial r} \approx -N\alpha I(r,z,t)r/r_0^2;\ N = p/k_B T$$

$$P_d = \frac{m^2(\xi - v_z)^2}{l_c k_B T} f_0(\xi - v_z)\Delta^4;$$

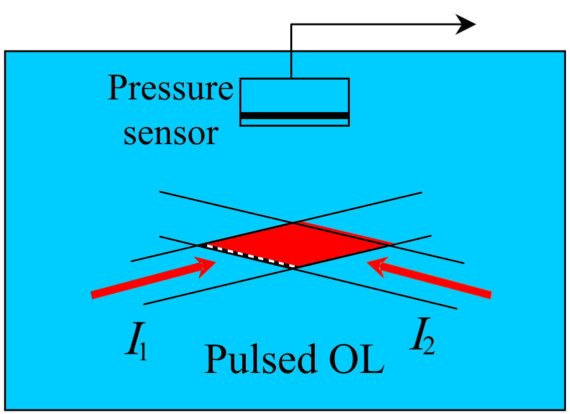
$$F_z = \frac{m^2(\xi - v_z)}{l_c k_B T} f_0(\xi - v_z)\Delta^4;$$

Formation of localized gas jets in “free space”

Air, p_0=1 Atm; T_0 = 300 K



Temperature and pressure distributions after the pulsed optical lattice



Proposed experiment: detection of the acoustic signal

Shneider, Barker, Submitted to Phys.Rev. A

Conclusions

•CRBS represents fast, non-intrusive laser diagnostic technique; Kinetic theory matches with experiment in wide range of gas densities

•An ensemble of molecules can be accelerated to high energies using an accelerating lattice created by the interference of two chirped laser beams

•Decelerating already cold molecules (~1 K) in a pulsed jet, creating a cold ensemble of stationary molecules at temperatures in the 1 mK range, appears feasible

•Such schemes are universal since all atoms and molecules are polarizable

•Energy and momentum exchange between the traveling OL wave and polarizable particles is analogous to the Landau damping of the electrostatic waves in a plasma

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