MOLECULAR ENSEMBLES
IN NON-RESONANT OPTICAL LATTICES

Mikhail N. Shneider

Applied Physics Group, MAE Department,
Princeton University, Princeton, NJ 08544

Physics Department, Friedrich-Schiller University, Jena
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Outline

- Optical lattice and Gradient Dipole Force
- Coherent Rayleigh-Brillouine scattering: *experiment and theory*
- Particle trapping, acceleration/deceleration and separation
- Optical manipulation with molecular beam: *experiment and theory*
- Non-resonant laser radiation absorption in the gas: Optical Landau Damping
- Optical Lattice – Gas Interaction (2 laser waves mixing): feed-back
- Conclusions
Optical Lattice

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 ($\beta$).

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

$\xi(t) = \frac{\Omega}{q}$

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

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

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Polarizability

• In general the polarizability tensor has a dispersive (Hermitian) and absorptive (non-Hermitian) part

\[ \alpha = \alpha' + i \alpha'' \]

• For an atom in its ground state \(|g\rangle\) the polarizability as a function of frequency given by the sum over all states

\[ \alpha(\omega) = \frac{2e^2}{3\hbar} \sum_l \frac{\omega_{l0} \left| \langle l | r | 0 \rangle \right|^2}{(\omega_{l0} - \omega - i\Gamma_l / 2)(\omega_{l0} + \omega + i\Gamma_l / 2)} \]

• At \(\omega \ll \omega_{l0}\), \(\alpha \neq \alpha(\omega)\)

• Polarizability is in general a tensor

\[
\begin{pmatrix}
  p_x \\
  p_y \\
  p_z
\end{pmatrix} = \begin{pmatrix}
  \alpha_{xx} & \alpha_{xy} & \alpha_{xz} \\
  \alpha_{yx} & \alpha_{yy} & \alpha_{yz} \\
  \alpha_{zx} & \alpha_{zy} & \alpha_{zz}
\end{pmatrix} \begin{pmatrix}
  E_x \\
  E_y \\
  E_z
\end{pmatrix}
\]
OL-gas interaction: standard theory limitations

- Gas is ideal
- Laser radiation frequencies are FAR from any resonance transition
- Laser pulse characteristics below breakdown
- No feedback from the gas particles to the laser field

For **fields far from resonance** the dipole optical potential and force on a particle of polarizability $\alpha$ are given by:

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

$$\vec{E} = \vec{E}_1 + \vec{E}_2$$

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

\[ F = -\nabla U = -\nabla \left( -\frac{1}{2} E \cdot \alpha \cdot E \right) \]

\[ F = \frac{1}{2} \alpha \nabla E^2 \]
Coherent Rayleigh-Brillouine scatteriωg (CRBS)

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

\[ \Delta \rho: \omega = \omega_1 - \omega_2 \]
\[ k = k_1 - k_2 \]

\[ I_s \propto L^2 \Delta n^2 I_1 I_2 I_3 \propto L^2 (\alpha \delta \rho)^2 I_1 I_2 I_3 \]

Works also for weakly ionized plasma, when \( \Delta n_{pl}(\omega) < \ll \Delta n_{gas}(\omega) \)

Positive column of glow discharge, flame, etc, where \( n_e / N < \ll 1 \)

with:
\[ \vec{F} = -\nabla U; \quad U = -\frac{1}{2} \alpha E^2; \quad E_a^2 = 2I / \varepsilon_0 c \]

\[ \delta \rho(x,t) = \int_{-\infty}^{\infty} \left[ f(v,x,t) - f_0(v) \right] dv \]
Coherent Rayleigh-Brillouin scattering:  
As example of four wave mixing in nonlinear optics

FIG. 11. Phase matching schemes for coherent Rayleigh-Brillouin scattering. $k_1$ and $k_2$ are the pump beams’ wave vectors. $k$ is the wave vector of the density perturbation.

CRBS in inert gas: Argon

\[ T_0 = 292 \text{ K} \]

\[ c_s(T) = \frac{\Delta \omega}{q} \]

Theory: Boltzmann eq. with the BGK collisional term

\[ y = \frac{1}{qv_0 \tau} \sim \frac{\lambda}{l_n}; \quad v_0 = \sqrt{\frac{2k_B T_0}{M}}; \quad q \approx \frac{4\pi}{\lambda} \]

CRBS in Molecular Gas

Theory based on the Wang-Chang-Uhlenbeck approach to kinetic equation with internal degrees of freedom:

\[
\frac{\partial f_i}{\partial t} + v \frac{\partial f_i}{\partial x} + \frac{F(x,t)}{m} \frac{\partial f_i}{\partial v} = \left( \frac{\partial f_i}{\partial t} \right)_c
\]

\( f_i \) is the space –velocity-time distribution function for molecules in state \( i \).

At equilibrium, \( f_i \) has a Gaussian distribution of velocities and a Boltzmann distribution of states.

\( N_2; T_0 = 292 \, \text{K} \)

Method for definition of the bulk viscosity \( \eta_b \) and rotational relaxation time, sound velocity, local gas temperature.
Example: measurement of sound velocity and gas temperature at low density

The same gas conditions: \( \text{N}_2, \ p=0.1 \ \text{Atm}, \ T=292 \ \text{K} \)

Pump laser beams wave length \( \lambda=532 \ \text{nm} \), but different angles \( \Theta \)

\( \Theta=180^0; \ y=0.59; \ \lambda_L=266 \ \text{nm} \)

Rayleigh shape: measurement of T

\( \Theta=5^0; \ y=1.35; \ \lambda_L=6100 \ \text{nm} \)

Rayleigh-Brillouine shape: measurement of \( C_s \) and \( \gamma=c_p/c_v \)
Arc discharge in an inert gas between the carbon electrodes

Idea: scattered signal from a pure gas is highly different from a gas with impurities: nanoclusters, buckyballs, nanotubes etc

Incoherent: Rayleigh, Rayleigh-Brillouine, etc

Coherent: Rayleigh, Rayleigh-Brillouine

Problems for diagnostics:
- Where clusters are formed
- Rate of formation
- Spectrum of size scales
- Nanotube alignment control
- etc

Laser scattering from this point (r,t): incoherent or coherent

Idea: scattered signal from a pure gas is highly different from a gas with impurities: nanoclusters, buckyballs, nanotubes etc

Coherent vs Incoherent Scattering

- For incoherent scattering the intensity increases as \( N \cdot N \) – number of scatterers
  - Scattered signal intensity decays as \( 1/r^2 \)

- For coherent scattering, the peak intensity is \( N^2 \) times the single dipole intensity
  - Scattered signal is a laser beam
    CRBS
Application of coherent Rayleigh-Brillouin scattering for \textit{in situ} nanoparticle and large molecule detection

M. N. Shneider$^{1,a)}$ and S. F. Gimelshein$^2$

$^1$Princeton University, D-429 Engineering Quad D-Wing, Princeton, New Jersey 08544, USA
$^2$University of Southern California, UPC, RRB 104, Los Angeles, California 90089-1191, USA

Example mixture: \(\text{Ar} + \text{C}_{60}\)
\(p_{\text{Ar}} = 1\text{ Atm}; T_0 = 300\text{ K}\)

Scattered CRBS signal:

\[ I_s \propto \Delta n^2 = \left( \sum_i \Delta n_i \right)^2 \]
\[ \Delta n_i = \frac{\alpha_i \Delta N_i}{2\varepsilon_0} \]

\(\checkmark\) Partial composition \(\text{Ar}:\text{C}_{60}\) up to \(10^{-5}\)
\(\checkmark\) Non-intrusive
\(\checkmark\) High time and spatial resolution
\(\checkmark\) Direct \(\text{C}_{60}\) density measurements

Computed CRBS signal in \(\text{Ar}:\text{C}_{60}\) mixture
Coherent Rayleigh scattering: (nonlinear regime: $U \leq kT$)

$\text{CO}_2; p=15 \text{ mBar}; T_0=300 \text{ K}$

$I_0=5 \times 10^{15} \text{ W/m}^2; U_{\text{max}}=39.3 \text{ K}$

$\tau_{\text{pulse}}=10 \text{ ns (FWHM)}$

Predicted: Shneider, Barker, Pan, Miles, Optical Commun., 239, 2004

Experimental confirmation: Bookey, Shneider, Barker, PRL 2007

Dicke, 1953: at elevated pressures – narrowing of a Doppler broadening

CRS: at reduced pressures – narrowing of CRS signal
Coherent Rayleigh scattering: mathematical model

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

\[ F = -\nabla U; \quad U = -\frac{\alpha}{\varepsilon_0 c} \sqrt{I_1(t)I_2(t)} \cos(qx - \Delta \omega t) \]

\[ \delta \rho(x, t) = \int_{-\infty}^{\infty} \left( f(v, x, t) - f_0(v) \right) dv \]

\[ I(\Delta \omega) \equiv I(\xi) = \frac{1}{\lambda} \int_{-\lambda/2}^{\lambda/2} \delta \rho(x, t)^2 dx \]

\[ \xi = \Delta \omega / q; \quad q = 4\pi / \lambda \]
Collisionless trapped and untrapped motion induced by Optical Lattice

\[ \ddot{x} = -a \cos(qx - \Omega t) \]

\[ \Omega = \omega_1 - \omega_2; \quad q = k_1 + k_2 = 4\pi / \lambda \]

\[ a = \frac{F}{m} = \frac{\alpha q E_0^2}{2m} = \frac{q U_{\text{max}}}{2m} \]

Phase: \( \theta = qx - \Omega t \)

New variables: \( x \rightarrow \theta; \quad t \rightarrow \tau = \sqrt{aqt} \)

Pendular motion: \( \ddot{\theta} = -\cos \theta \)

Phase plain (\( \theta, \eta \)): \( \dot{\theta} = \eta \)

\( \dot{\eta} = -\cos \theta \)
Optical Landau damping and induced drift

\[ \lambda I_c \sim 1 \]

\[ \Omega = \omega_2 - \omega_1 = \text{const}; \quad \zeta = \Omega/q = \text{const}; \]

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

\[ \frac{\partial f}{\partial t} + v \frac{\partial f}{\partial x} + \frac{m}{m} \frac{\partial f}{\partial v} = - \frac{f - f_0}{\tau} \]

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.

Example: Ar; \( p = 1.33 \text{ Pa}; \ T = 300 \text{ K}; \ \lambda = 800 \text{ nm} \]

\( l = 1.6 \times 10^{16} \text{ W/m}^2; \ U = 78 \text{ K}, \ z = 316 \text{ m/s} \)

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

Drift: \( v_{dr} (\zeta) = \left\langle \frac{1}{N} \int_{-\infty}^{\infty} f(x,v,t)vdv \right\rangle \approx 5 \text{ m/s} \)

Acceleration of an ensemble in chirped optical lattice

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

\[ \left( \frac{\partial f}{\partial t} \right)_c = 0 \]

\( CH_4; \ p = 0.1 \text{ Torr}; \ T_0 = 300 \text{ K.} \)

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

For this calculation:
\( \psi = 0.59, \ q = 1.57 \times 10^7 \text{ m}^{-1}, \)
\( a = 2.14 \times 10^{12} \text{ m/s}^2, \ \Delta U = 133 \text{ K}, \)
\( I = 3 \times 10^{12} \text{ W/cm}^2. \)
\( \beta = \frac{d\omega}{dt} = 10^{19} \text{ rad/sec}^2. \)

From “lasers + gas” conservation of momentum and energy:
more than 10^9 molecules can be trapped and accelerated

Collisional acceleration of CH$_4$ (Gaussian pulse)

DSMC calculations

CH$_4$; p=3 Torr; $T_0$=300 K
Gauss pulse in time

CH$_4$; p=10 Torr; $T_0$=300 K
Gauss pulse in time


DSMC computational tool used: **SMILE** (Ivanov, Markelov, Gimelshein AIAA Paper 98-2669)
Example: Pulsed optical lattice interacting with \((C_{60} + \text{He})\) molecular beam

\[
\frac{\alpha/M}{C_{60}} \approx 1.8
\]

Selective acceleration of \(C_{60}\), hit the surface, \(Mv^2/2\), \((p,T)\) optimal for diamond

Separation based on $\alpha/M$ difference

Application of time varying lattice forces, created by controlling the intensity and therefore well depth, could be used to induce a temporal velocity dispersion based on the polarisability to mass ratio in a multi-component mixture.

**Molecular beam:** $^{30}\text{N}_2:^{28}\text{N}_2:\text{CO} = 1:1:1; \quad v_b=239.1 \text{ m/s}; \quad T=5 \text{ K}

**Optical lattice:** $I=3.3 \times 10^{11} \text{ W/cm}^2$;

$\beta=5.47 \times 10^{17} \text{ rad/sec}^2$

\[\psi > 1; \quad \Delta t=5 \text{ ns}\]

G. Dong, W. Lu, P. F. Barker, M.N.Shneider, Progress Quant. Electronics, 29 2005
Figure 9. Five resolved isotopes in the time-of-flight spectrum (bold) of the unperturbed xenon in the molecular beam. Also shown (filled grey distribution) is the time-of-flight spectrum of decelerated xenon atoms in a 373 m s$^{-1}$ lattice.
Cold (<1 K) and Ultracold (<10^{-3} K) Molecules

Importance:
- High resolution spectroscopy
- Collision and Van der Waals interactions studies
- Cold chemistry:

Rate of chemical reactions (Arrhenius law): $K_A \sim \exp(-E/kT)$

At $T \leq T_{tun} = (\sqrt{2\hbar/\pi kd})\sqrt{E/M}$, $K_{tun} > K_A$ (Hund, 1927; Gol’danskii, 1959)

$T_{tun} < 10 \text{ K}$
Cold (<1 K) and Ultracold (<10^{-3} K) Molecules

Fig. 1. Examples of temperature dependence of the rate constant for the reactions in which the low-temperature rate-constant limit has been observed: 1. hydrogen transfer in the excited singlet state of the molecule represented by (6.16); 2. molecular reorientation in methane crystal; 3. internal rotation of CH_3 group in radical (6.25); 4. inversion of radical (6.40); 5. hydrogen transfer in "halved" molecule (6.16); 6. isomerization of molecule (6.17) in excited triplet state; 7. tautomerization in the ground state of 7-azoindole dimer (6.1); 8. polymerization of formaldehyde in reaction (6.44); 9. limiting stage (6.45) of (a) chain hydrobromination, (b) chlorination and (c) bromination of ethylene; 10. isomerization of radical (6.18); 11. abstraction of H atom by methyl radical from methanol matrix [reaction (6.19)]; 12. radical pair isomerization in dimethylglyoxime crystals [Toriyama et al. 1977].

V.A. Benderskii, V.I. Goldanskii, D.E. Makarov, Physics Reports, 1993
Cold (<1 K) and Ultracold (<10^{-3} K) Molecules

Methods:

• Photoassociation of ultracold atoms
  Stwalley’s group (University of Connecticut); Fioretti et al (Piza, Italy)

• Buffer gas cooling - dissipation provided by thermalisation with cold gases - 100 s mK
  Doyle’s group (Harvard)

• Cooling in a supersonic expansion + deceleration
  - Electrostatic slowing of polar molecules - G. Meijer’s group (Berlin) and others (200 mK)
  - Slowing polarizable species in optical lattices
Molecular beam trapping and slowing down

\[ H(x) = 0.5u^2 + c_p T(x) = c_p T_0 \]

At \( x/D > 5 \):

\[ v_b \sim u_\infty = c_0 [2/(\gamma - 1)]^{1/2} = [2(\gamma R T_0) / (\gamma - 1)]^{1/2} \]

\[ \theta \approx 180^\circ \]

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

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

\[ q = |\vec{k}_1 - \vec{k}_2| \approx 2k_1 \]

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.

The deceleration of an ensemble of particles

Boltzmann equation:

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

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

Iodine

I = 1.2 x 10^{14} \text{ W/m}^2; \ \psi = 0.735

v_0 = 560 \text{ m/s}; T_0 = 1 \text{ K}

I_2: U_{max} = \alpha I_2z_\theta = 1 \text{ K}

Ar: U_{max} = 0.157 \text{ K}

\(\delta N_{Ar}/N_{0, Ar} = 1.6 \%\)

\(\delta N_{I_2}/N_{0, I_2} = 8 \%\)

Controlling the motion of cold molecules with deep periodic optical potentials

R. Fulton1, A. I. Bishop1, P. F. Barker1 and M. N. Schneider2

1Physics, School of Engineering and Physical Sciences, Heriot Watt University, Edinburgh EH14 4AS, UK
2Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544, USA

NO; \( v_b = 400 \text{ m/s} \); \( T_0 = 1.8 \text{ K} \)

Figure 1 The orientation of the cold NO molecular beam with respect to the pulsed optical lattice. The ultraviolet probe beam that was used to ionize and detect the NO molecules perturbed by the lattice is also shown. The velocity of the ions, and thus that of the neutral molecules, was determined from their time-of-flight in a mass spectrometer.

Figure 3 The velocity distribution function of decelerated NO molecules calculated as a function of time by numerical integration of the one-dimensional Boltzmann equation. a. The temporal lattice intensity profile produced by separate measurements of the two Q-switched Nd:YAG laser profiles used to create the optical lattice. b. The simulation of the dynamics of molecules within a constant-velocity optical lattice using the lattice profile of a. A 321 m s⁻¹ lattice with an average well depth of 22 K was used to decelerate a portion of NO molecules from 400 to 270 m s⁻¹ representing a near-half rotation in phase space.

Figure 4 The velocity distribution function of NO molecules perturbed by the optical lattice. Lattice velocities with respect to the molecular-beam velocity of -79, -52, -26, 0, 26 and 52 m s⁻¹ are shown. For comparison, simulations of the perturbed distribution function created by the different velocity lattices is shown alongside the experimental data.
Pulsed optical lattice: benzene (C$_6$H$_6$)

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

\[ F = -\nabla U \]

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

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

\[ T_0 = 2.3 \text{ K}; \ v_b = 320 \text{ m/s} \]

Measured laser beam intensities.

Energy per pulse: $\Sigma_1 \approx \Sigma_2 \approx 0.345 \text{ J}$

After interaction with pulsed OL

Coherent Rayleigh and Rayleigh-Brillouine scattering in gases
• Gas and weakly ionized plasma (flame) diagnostics

"Linear" approach (no laser-gas feed-back) is valid!

“Non-Linearity” and Feed-back is important:

Pulsed and continuous Optical Lattices
• Acceleration/deseleration
• Transport
• Separation: $\alpha_i/M_i$
• Power deposition
• Microscale gas mixing

Molecular transport in pulsed optical lattices

1 Princeton University, Engineering Quad., Olden Street, Princeton, NJ 08544, USA
2 University College London, London WC1E 6BT, UK
3 University of Southern California, Los Angeles, CA 90089, USA
OL-gas interaction: theory limitations

- Gas is ideal
- Laser radiation frequencies are FAR from any resonance transition
- Laser pulse characteristics below breakdown
- Feed back from the gas particles to the laser field
How does feedback happen?

Optical Lattice → ΔN → Δn → Bragg reflection → Optical Lattice

ΔN – gas density perturbation; Δn – perturbation of the index of refraction

Challenges for self-consistent theory: Very weak interaction, because of Δn/(n_0-1)<<1; Typical optical lattice scale ~1000’s of λ
Electromagnetic wave in a media:
\[
\frac{\partial^2 E}{\partial x^2} - \frac{n^2}{c^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{nl}}{\partial t^2}
\]

The refractive index is \( n = n_0 + \Delta n(x,t); \ |\Delta n| \ll n_0 \)

\[
n = \sqrt{1 + \frac{N \alpha}{\varepsilon_0}} \approx n_0 + \Delta n(x,t); \quad n_0 = 1 + \frac{N_0 \alpha}{2 \varepsilon_0}
\]

\[
\frac{\partial^2 E}{\partial x^2} - \frac{n_0^2}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{2n_0 \Delta n}{c^2} \frac{\partial^2 E}{\partial t^2}
\]

\[
\Delta n(x,t) = \frac{\Delta N(x,t) \alpha}{2 \varepsilon_0}
\]

\[
\Delta N(x,t) = \int_{-\infty}^{\infty} [f(x,v,t) - f_0] dv
\]

\[
\frac{\partial f}{\partial t} + v \frac{\partial f}{\partial x} + \frac{F(x,t)}{m} \frac{\partial f}{\partial v} = -\frac{f - f_0}{\tau_c}
\]

\[(1) \quad E = E_1 + E_2\]

(2) Perturbation of index of refraction

(3) Gas density perturbation

(4) Boltzmann kinetic equation in BGK

\[
F(x,t) = -\frac{\partial}{\partial x} U = \alpha \left( \frac{\partial E_1}{\partial x} E_2 + \frac{\partial E_2}{\partial x} E_1 \right) \cos(qx - \Omega t) - \frac{\alpha}{2} E_1 E_2 q \sin(qx - \Omega t)
\]

\[
U(x,t) = -\frac{1}{2} \alpha E^2(x,t)
\]

Shneider, Barker, Optics Commun., 284, 2011
Conservation of energy:
\[
\frac{\partial W_E}{\partial t} + \nabla \cdot \Pi = -\frac{\partial W_g}{\partial t}
\]

Density of Electromagnetic Energy:
\[
W_E = \left\langle \frac{\varepsilon_0 \varepsilon (E_1 + E_2)^2 + \mu_0 \mu (H_1 + H_2)^2}{2} \right\rangle_{2\pi/\omega}
\]

Poynting vector (flux of Electromagnetic energy):
\[
\Pi_i = \frac{\varepsilon_0 c}{2} \sqrt{\frac{\varepsilon}{\mu}} E_i^2 k_i = I_i k_i
\]

\[
I_1(0) - I_1(L) + I_2(L) - I_2(0) = \int_0^L [\int F(x,t)f(x,v,t)v dv] dx
\]

Conservation of momentum

In a general case:
\[
\frac{d}{dt} (\Xi_{gas} + \Xi_{field})_x = \oint_S T_{xn} dS
\]

\(\Xi_{gas}\) - momentum of gas in the interaction region
\(T_{ik} = \varepsilon_0 (E_i D_k + H_i B_k) - \frac{1}{2} \delta_{ik} \varepsilon_0 (\vec{E}\vec{D} + \vec{H}\vec{B})\) - Maxwell stress tensor

For a pulsed Optical Lattice:
\[
\Xi_{gas,x} = \int_0^\tau \int_0^\infty \left\langle T_{xx} >_{2\pi/\omega} \right\rangle dS_x dt = \frac{S_x}{c} \int_0^\tau [I_1(0,t) - I_1(L,t) - I_2(L,t) + I_2(0,t)] dt
\]
Examples

CH$_4$; $p=1$ Atm; $T=300$ K; $I_1(0)=I_2(L)=10^{16}$ W/m$^2$; $\omega_1 = \omega_2$; $L=200\lambda$

$I_1(x)/I_1(0), I_2(x)/I_2(L)$

$\Delta n_a = 3.7 \times 10^{-5}$

Shneider, Barker, Optics Commun., 284, 2011
Examples: “Amplifier”

CH$_4$; p=1 Atm; T=293 K; $I_1(0)=10^{16} \gg I_2(L)=10^{12}$ W/m$^2$;

$\omega_1 = \omega_2$; $L=200\lambda$; $\lambda=532$ nm

$I_1(0) \gg I_2(L)$

At $L=1$ cm$\approx 2 \times 10^4 \lambda$

Gain=$I_2(L)/I_2(0) \approx 6.4$

Effect as stronger as $I_2(L) \ll I_1(0)$

$\Delta n_a = 3.56 \times 10^{-7}$

$I_1(0)=10^{16}$ W/m$^2$

$I_2(L)=10^{12}$ W/m$^2$

$L=200\lambda$; $\xi=0$

Shneider, Barker, Optics Commun., 284, 2011
Application to a remote atmosphere laser “amplifying”

1. Remote Oxygen or Nitrogen laser: *Intensity is low*
   
   Dogariu et al, Science 2012; Kartashov et al, PRA 2012

2. Superimposed with “pump upward” laser beam of the same wave length

3. Formation of the remote interference pattern

4. “Amplifying” of the “backward” laser beam
Experiment (Taylor Lilly et al, University of Colorado at Colorado Springs, 2013) versus theory: $N_2$; $p=0.8$ Atm; $T_0=293$ K; Nd:YAG laser; $\lambda=532$ nm

If $\lambda/l_c >> 1$ no plateau

Potential well mowing with phase velocity, $\xi$

$E_1 = 2\pi \int_0^\infty \int_0^\infty I_1(L, r, t) rdr dt$

$E_2 = 2\pi \int_0^\infty \int_0^\infty I_2(0, r, t) rdr dt$

**FIG. 3.** Comparison of post-interaction experimental energy difference with numerical prediction versus laser frequency difference.
Conclusions

• Powerful non-intrusive diagnostics for gas and gas-nanoparticle mixtures

• An excellent examples that classical problems in modern optics are still in force

• Kinetic theory matches with experiment in wide range of gas densities and laser intensities

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

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

• Many of potential technological applications:
  ➢ neutral molecular beams: etching and deposition; molecular-surface scattering studies
  ➢ separation in gas mixtures
  ➢ microscale mixing
  ➢ etc.

Further studies

• Optical lattices at critical regimes (when breakdown starts, etc)
  • …
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