Two Beam X-CARS in Dispersive Media

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CARS Thermometry

- Coherent Anti-Stokes Raman Spectroscopy (or scattering)
  - spatially resolved through crossed beams
  - instantaneous with pulsed lasers
  - high noise rejection potential with coherent signal beam

- Alignment geometries include BOXCARS (folded and planar), USED CARS, collinear CARS
Coherent Anti-Stokes Raman Spectroscopy -- Energy

\[ 2\omega_1 - \omega_2 = \omega_3 \]
CARS -- Temperature Fitting

T = 1016 K
Folded BOXCARS for Spatial Resolution

aligned with flame sheet

across flame sheet
CARS Measurements in a Wolfhard-Parker Burner
CARS Measurements in a Droplet Stream Flame

Methanol droplet stream flame
Typical Folded BOXCARS Experiment Apparatus
Geometry of BOXCARS

• $\theta$ is vertical angle between Stokes probe beam and the optical axis

• $\alpha$ is the horizontal half angle between the pump beams

• $\phi$ is the vertical angle that the CARS signal beam makes with the optical axis
CARS Signal Intensity

\[ I_3 = \frac{\omega_3^2}{n_1^2 n_2 n_3 c^4 \epsilon_0^2} I_1^2 I_2 |\chi_{CARS}|^2 l^2 \left( \frac{\sin \frac{\Delta kl}{2}}{\frac{\Delta kl}{2}} \right)^2 \]

- Signal depends on:
  - interaction length squared
  - third-order non-linear susceptibility squared
  - intensity of pump and Stokes probe beams
  - phase matching function
BOXCARS Phase Matching

\[ \Delta \vec{k} = 2\vec{k}_1 - \vec{k}_2 - \vec{k}_3 \]
Assume the $y$ phase match is automatically satisfied since the CARS signal beam selects its angle to do so.

Solving the $z$ phase match for zero then links $\alpha$ and $\theta$.

Ideal phase matching will depend on dispersion.

For $\alpha = 0$, only one pump beam is needed.
Note: for dispersionless media, the energy constraint forces collinear phase matching
Phase Matching Constraints

- Collinear CARS in air (dispersionless media) satisfies ideal phase matching.
- Folded BOXCARS ideal phase matching in both air and glass cannot be achieved simultaneously.
- CARS in dispersionless media cannot be achieved with 2 crossing beams.
- CARS in dispersive media can be achieved with 2 crossing beams (X-CARS).
- Folded BOXCARS and twin X-CARS ideal phase matching cannot simultaneously be achieved in glass.
Observed CARS Signals from Glass

\[ \alpha = 1.2 \text{ degrees} \]
\[ \theta = 2.8 \text{ degrees} \]

Twin X-CARS signals

When Stokes and two pump beams all overlap, 3 beam BOXCARS
Dreier and co-workers have a working geometry consistent with X-CARS (planar BOXCARS with 0.95 degrees pump half-angle at pressures to 2500 atm)
experimentally observed simultaneous twin X-CARS in glass and BOXCARS in air and glass
Phase Mis-matched BOXCARS

\[ I_3 = \frac{\omega_3^2}{n_1^2 n_2 n_3 c^4 \varepsilon_0^2} I_1^2 I_2 |\chi_{CARS}|^2 l^2 \left( \frac{\sin \frac{\Delta kl}{2}}{\frac{\Delta kl}{2}} \right)^2 \]

\[ \Delta k_z = 2 \omega_1 n_1 \cos(\alpha) - \omega_2 n_2 \cos(\theta) - \omega_3 n_3 \cos(\phi) \]

Phase match assumed ideal, as before

\[ l \] is controlled either by the geometry of the beam crossing (thick media) or by the media thickness (thin media)
CARS Signal vs. Phase Match

Twin XCARS
Ideal phase match
In glass

folded BOXCARS
Ideal phase match
In air

Folded BOXCARS
Ideal phase match
In glass

Pump angle in AIR (degrees)

Stokes angle in AIR (degrees)
Constant Interaction Length
CARS Signal Strength
fixed interaction length

![Graph showing signal strength as a function of pump and Stokes angles in AIR degrees. The graph vividly illustrates the signal intensity in arbitrary units across different angles.]
Conclusions

● Except for the collinear configuration, ideal CARS phase matching with two beams in non-dispersive media is not possible;

● Ideal CARS phase matching with two crossed beams in dispersive media is possible; X-CARS common in condensed phase measurements;

● Ideal folded BOXCARS phase matching conditions simultaneously in air and glass is not possible;

● Because the CARS signal strength depends on interaction length and phase matching, strong CARS signal far from ideal phase matching with appropriate interaction lengths is possible;

● With the proper geometry, it is possible to create a system that can generate a BOXCARS signal in glass, a BOXCARS signal in air, and twin X-CARS signals in glass; such a geometry may be useful to aid alignment in the system;

● The potential for X-CARS in dispersive media may allow a simpler spatially resolved CARS measurement in high pressure combustion environments than is typical.
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