UV-VIS AND POLARIZATION RESOLVED
LIGHT SCATTERING OF MIXED PHASES
AEROSOL PARTICLES

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General concern:
Interaction of a laser beam with the atmosphere

1. Coupling pulsed laser pulse with Lidar technology
   It allows range resolved observation of the atmosphere content (0-10 km range, range resolution = 10 m).

2. Performing observation in the real thermodynamic conditions.

3. The interaction is sensitive to traces, particles, clouds,…


> 100’000 citations (laser atmosphere remote sensing)

What is new?

i) Improvement of the observation quality: accuracy and reliability, novel species.

ii) Rethinking photometry theory and light detection scheme.

Left: Laser beam into the atmosphere, laser wavelength = 532 nm. Right: Range corrected Lidar signal.
Spectra of the atmosphere backscattering from a air parcel at an altitude of 300 m above ground level in Lyon.
Main concern: remote sensing of laser light backscattering of atmospheric particles

Optical properties of particles:

- **Backscattering**
  - Sensitive to particle size and shape (Mishchenko et al., *Cambridge Univ. Press*, 2002);

Light backscattering: light scattering with $\theta = 180^\circ$ ($k_{\text{inc}} = -k_{\text{sca}}$)

- **Polarization**

Particle nonsphericity (Mishchenko et al., *J. Quant. Spec. Rad. Transf.*, 2009)

$I_{\text{inc}}$, $I_{\text{sca}}$ polarization with respect to the laser linear polarization

$$\delta_p = \frac{I_{\text{sca},\perp}}{I_{\text{sca},//}}$$

$\delta_p$: particle linear depolarization ratio

- **Wavelength**

Particle size (Sasano and Browell, *Appl. Opt.*, 1989). (Mishchenko et al., JQRST 2013);
1. Aerosol particles in the atmosphere: shape, size, optical properties.

2. Optical partitioning in two/three component particles of atmospheric aerosol.

3. Laboratory measurement of particles scattering at the exact backscattering angle.

4. Conclusion
Hypothesis: Ensemble of spherical or non-spherical particles externally mixed and suspended in the air.

How light scattering is sensitive to such a complexity?

How to perform reliable observation?

\[ (p) = \text{ns1} + \text{ns2} + \text{n12} \]

TEM = Transmission Electron Microscope.
Numerical simulation of particles Backscattering cross-sections

T-matrix simulation, 355 nm (blue line), 532 nm (green line)

G. David et al., ACP (2013)
Numerical simulation of Backscattering cross-sections averaged over the particle size distribution

Refractive index, $m = 1.57 - 0.007i$ (Kandler et al., 2011)
Aspect ratio $\varepsilon$: 1.2, 1.4, 1.6,..., 2.6, Merikallio et al., ACP, (2011).

G. David et al., ACP (2013)
**Physical Nature of the Measurement**

**General frame and assumptions**

- Ensemble of randomly-oriented particles
- Single-scattering approximation
- Far-field approximation
  
  (Mishchenko et al., *Cambridge Univ. Press*, 2002)

**Scattering matrix \( \mathbf{F} \)**

In the exact backscattering geometry,

\[
\begin{pmatrix}
I_{sca} \\
Q_{sca} \\
U_{sca} \\
V_{sca}
\end{pmatrix}
= \frac{C_{sca,p}}{4\pi d^2}
\begin{pmatrix}
F_{11,p} & 0 & 0 & F_{14,p} \\
0 & F_{22,p} & 0 & 0 \\
0 & 0 & -F_{22,p} & 0 \\
F_{14,p} & 0 & 0 & F_{11,p} - 2F_{22,p}
\end{pmatrix}
\begin{pmatrix}
I_{inc} \\
Q_{inc} \\
U_{inc} \\
V_{inc}
\end{pmatrix}
\]

- \( p \) = particle
- \( C_{sca,p} \) : particle scattering cross-section
- \( d \) : distance from the scattering volume to the observation point.

\( F_{ij} \) : scattering matrix elements

wavelength, size, shape, refractive index-dependent.
1. Aerosol particles in the atmosphere.  
   Shape, size, optical properties

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4. Conclusion and perspectives
UV-VIS polarization Lidar detector

- Home-built UV-VIS polarization lidar detector
  \[ \lambda = 355 \text{ nm (UV)} \text{ and } 532 \text{ nm (VIS)} \]

- Cross talk
  \[
  \begin{pmatrix}
  I^\perp,\lambda^* \\
  I^\perp,\lambda \\
  I^\perp,\lambda^* \\
  I^\perp,\lambda
  \end{pmatrix}
  =
  \begin{pmatrix}
  \eta^\perp,UV & 0 & 0 & 0 \\
  3.8 \times 10^{-8} \eta^\perp,UV & 0 & 0 & 0 \\
  0 & 0 & \eta^\perp,VIS & 0 \\
  0 & 0 & 0 & \eta^\perp,VIS
  \end{pmatrix}
  \begin{pmatrix}
  I^\perp,UV \\
  I^\perp,UV \\
  I^\perp,VIS \\
  I^\perp,VIS
  \end{pmatrix}
  \]

- Calibration
  Following the methodology of J. Alvarez et al., 
  \[ G_{UV} = 29.16 \pm 0.22 \]
  \[ G_{VIS} = 16.69 \pm 0.23 \]

Negligible cross-talk between \{//,\perp\} polarization, \{UV-VIS\} wavelength channels (better than $10^{-7}$ accuracy)
Multi wavelength Polarization-Sensitive Detector of backscattering radiation

355 nm Polarization Channels

532 nm Polarization Channels

607 nm Raman N$_2$ Channel

408 nm Raman H$_2$O Channel

660 nm Raman H$_2$O Channel

Input
Polarized resolved UV-VIS particles backscattering

October 18th 2011 at Lyon (France)

\[ \beta_{p,\perp} \text{(VIS)} \]

\[ \beta_{p,\perp} \text{(UV)} \]

Fig. 6 Cross-polarized UV-particles backscattering time-altitude map

Fig. 7 Cross-polarized VIS-particles backscattering time-altitude map

\{\parallel, \perp\} are defined with respect to laser linear polarization.
Interpreting lidar returns using scattering matrix formalism

- Case of a two-component particles mixture \((p) = \{s, ns\}\)

\[
\beta_p = \beta_s + \beta_{ns}
\]

\[
\delta_p < \delta_{ns}
\]

\[
\Rightarrow \delta_p \text{ is not a quantitative indicator of ns particles content}
\]

How to partition the light scattered by a particles mixture in its \(\{s, ns\}\) components

4 unknown quantities namely \(\beta_{p,\pi}\) with \(p = (s, ns)\) and \(\pi = \{/, \perp\}\)

\[
\beta_{p,\perp} = \beta_{ns,\perp}
\]

\[
\beta_{p,\perp} = \beta_{ns,\perp}
\]

\[
\beta_{ns} = \beta_{ns,\perp} + \beta_{ns,\perp} = \beta_{ns,\perp} (1 + 1/ \delta_{ns})
\]

Value of \(\delta_{ns}\) is required:

- Laboratory measurements (O. Muñoz et al.,)
- Numerical simulations (T-matrix, Mishchenko code, Kahnert code, DDA-code, T. Nousiainen)

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1: A. Miffre et al., GRL, 2011.
Particles backscattering at Lyon
(Eyjafjallajökull volcanic eruption, April 2010)

Eyjafjallajökull Icelandic volcano eruption
(MODIS satellite, NASA, April 19th 2010).

Ash particles numerical dispersion model at Lyon (A. Stohl’s group).

Volcanic ash particles number concentration retrieval

- Ash particle refractive index at 355 nm:
  \[ m = 1.54 \pm 0.0054i \]  (P. Winchester, 1998)

- Ash particle size distribution:

\[
N_{\text{ash}}(z) = \beta_{\text{ash}}(z) \left/ \langle \frac{d\sigma}{d\Omega} \rangle_{\text{ash}} \right. 
\]

- Ash particle sedimentation:

Fig. 8 Literature references on ash particles size distribution (PSD).

- \( C_{\text{sca}} \) (Muñoz, 2004, Different volcanoes) = 16.4 \( \times 10^{-9} \) cm\(^2\) part\(^{-1}\)
- \( C_{\text{sca}} \) (Ilyinskaya, 2011, Eyjafjallajökull, Iceland) = 22.2 \( \times 10^{-9} \) cm\(^2\) part\(^{-1}\)
- \( C_{\text{sca}} \) (Schumann, 2011, Eyjafjallajökull, Germany) = 17.0 \( \times 10^{-9} \) cm\(^2\) part\(^{-1}\)

UV-polarization backscattering during desert dust outbreak: NPF

Field measurements have the same behavior that Laboratory experiment on new particle formation (NPF) C. George’s group,¹ and H. Hermann ground based measurements¹

Case of a three-component particles mixture \((p) = \{\text{ns1, ns2, n12}\}\)

\[
\begin{align*}
(p) &= \text{ns1} + \text{ns2} + \text{n12} \\
\beta_{p,\pi}(\lambda) &= \{/\, \perp\} \\
\lambda &= \{\text{UV, VIS}\}
\end{align*}
\]

How to partition the mixture in its three \(\{\text{ns1, ns2, n12}\}\) components\(^1\)

- UV-VIS polarization lidar: \(\beta_{p,//}(\text{UV, VIS}), \beta_{p,\perp}(\text{UV, VIS})\) 
- Numerical simulations in collaboration with T. Nousiainen

Dust particles (ns1) : T-matrix code (M.I. Mishchenko, 1998) 
Sea-salt particles (ns2) : Extension of T-matrix code for cubes (M. Kahnert, 2013).

Model Inputs:
Particles mixture = ensemble of spheroids \((r, \epsilon, \lambda, m)\), PSD of ns1, ns2-particles

Model Outputs:
UV-VIS depolarization ratios : \(\delta_{\text{ns1}}, \delta_{\text{ns2}}, \delta_{\text{n12}}\)
Cross-polarized Angstrom exponent \(A_{\text{ns},\perp}\) for ns1, ns2-particles

\(^1\) G. David et al., ACP(2013).
Retrieved backscattering profiles of a three-component particle mixture\(^1\)

UV-VIS backscattering vertical profiles (20% uncertainty) of dust, sea-salt (ss) and spherical (s) particles in the three components mixture. October 18\(^{th}\) 2011 at 16h15 UTC at Lyon (France)

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Outline

1. Aerosol particles in the atmosphere.
   Shape, size, optical properties

2. Optical partitioning in two/three component particles of atmospheric aerosol.

3. Laboratory measurement of particles scattering at the exact backscattering angle

4. Conclusion and perspectives
Laboratory experiment at the exact Backscattering angle $^{1,2}$

Motivations

a. Need to provide insights on particles nonsphericity
   - For ns-particles: Determine the particles depolarization ratio $d_p$

b. Comparison with numerical simulations

c. Inverting remote sensing measurements: satellite-based or ground-based $^{2,3}$.

Methodology

Laboratory measurement: Polarization-resolved laser-light backscattering on an ensemble of particles in ambient air, at the exact backscattering angle $^1$.

Laboratory measurement at exact backscattering angle

Detector

PBC

QWP

Air surrounding medium

Backscattered pulse

Pulsed laser source

Particles

QWP

Incident pulse

D = 5 m

$$S_p(\psi) = \begin{bmatrix} 1 & 1 & -1 & 0 & 0 \\ 0 & -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & \cos^2(2\psi) & \sin(4\psi) / 2 & -\sin(2\psi) & 0 \\ 0 & \sin(4\psi) / 2 & \sin^2(2\psi) & \cos(2\psi) & 0 \\ 0 & \sin(2\psi) & -\cos(2\psi) & 0 & 0 \end{bmatrix} \begin{bmatrix} F_{11,p} & 0 & 0 & F_{14,p} \\ 0 & F_{22,p} & 0 & 0 \\ 0 & 0 & 0 & F_{22,p} \\ 0 & 0 & 0 & F_{11,p} - 2F_{22,p} \end{bmatrix} \begin{bmatrix} 1 \\ 0 \\ 0 \\ 0 \end{bmatrix}$$

Detected scattering angles $\theta = (180.0 \pm 0.2)^\circ$
Absolute depolarization ratio measurement of an ensemble of particles

\[ S_p(\psi) = \frac{\alpha}{(2D^2)} \times [F_{11,p} + F_{22,p} + (F_{11,p} - 3F_{22,p}) \times \cos (4\psi)] \]

Backscattering signal \( S_p \) (in black) as function of the orientation of the quarter-wave plate \( \psi \).

Left: water droplet, right: NaCl particle

\[ \delta_p = \frac{(F_{11,p} - F_{22,p})}{(F_{11,p} + F_{22,p})} \]

G. David, B. Thomas, E. Coillet, A. Miffre and P. Rairoux, Opt Exp. 2014, (June Spotlight)
Case study of NaCl particles

Depolarization measurements

RH = 90 %
\(d_{\text{Salt}} = (2.3 \pm 0.5) \times 10^{-2}\)

RH = 52 %
\(d_{\text{Salt}} = (11.3 \pm 1.4) \times 10^{-2}\)

RH = 30 %
\(d_{\text{Salt}} = (17.6 \pm 1.2) \times 10^{-2}\)

\(d_{\text{Salt}} \) (T-matrix) = 20.6 \times 10^{-2}

Extension of T-matrix for cubes (Kahnert, 2013)

\(S_{p, \tau}(\psi) = \alpha \left[ F_{11, p} + F_{22, p} + (F_{11, p} - 3F_{22, p}) \times \cos(4\psi) \right] \)

\(S_{p, \tau} \) : Signal \( S_p \) integrated over the laser pulse duration
\(\psi \) : QWP orientation angle
Conclusion

- Optical partitioning of a two/three component particles mixture is feasible by coupling 2\(\lambda\)-polarization lidar with an additional tool (laboratory measurement, numerical simulations).

- New and robust methodology, whose accuracy depends on the accuracy of both the lidar retrievals and single-scattering simulations. Need however to be validated by independent co-located measurements.

- Laboratory measurements of particles scattering at the exact backscattering angle is achievable and with high precision.
  - the complete phase function complex mixture (external, internal, ...).
  - model...
Coupling backscattering with the AIDA chamber

- Forthcoming experiments (Dec 2014) in collaboration with Dr. M. Schnaiter, IMK-AAF, KIT
Team work

Linked publications
2. G. David et al., OE, (2013).

Available CNRS’s
Post-doctoral Position

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Thank you for your attention