(Kandinski, 1926)

FRACTAL DUST PARTICLES: LIGHT SCATTERING AND ADSORPTION ANOMALIES

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(Kandinski, 1926)

ALMOST-KNOWN KNOWNS ABOUT FRACTAL DUST PARTICLES LIGHT SCATTERING IN VARIOUS DOMAINS OF PHYSICS

(AND POSSIBLE ASTROPHYSICAL APPLICATIONS)

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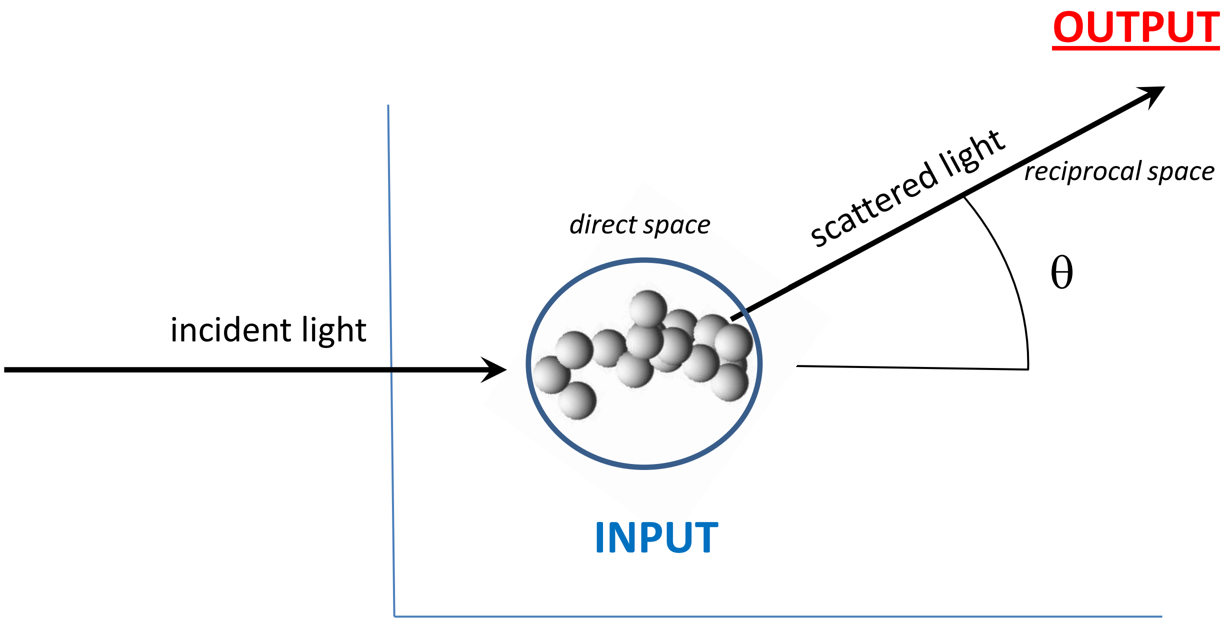
WHY IS THE INVERSE SCATTERING PROBLEM SO DIFFICULT?

- ***direct problem*** → scattering is computed solving the Maxwell equations with the correct limit conditions



INPUT = scatterer

OUTPUT = 4 functions, e.g.: Stokes parameters $S_i(\theta, \lambda)$

depend on a number of independent parameters
(θ, λ, m, distribution of the matter in the scatterer, size-distribution)

THE FREE NUMERICAL PROGRAMS TO DO SO…

Thank you for the support of our onlinesurvey.
Now we evaluate the results.

PROGRAMS

List of electromagnetic scattering programs

3D GLMT

Mie Type Codes

Written by Gouesbet, Gérard

Monday, 07 November 2011 13:34

3D representation of the scattering of a Gaussian beam by a homogeneous spherical particle based on the far-field version of GLMT to visualize the scattering of a Gaussian beam by a homogeneous, isotropic and spherical particle arbitrarily located in the beam. Supplementary resources with the book:

Gérard Gouesbet and Gérard Gréhan. Generalized Lorenz-Mie Theories. Springer, Berlin Heidelberg, 2011, ISBN 978-3-642-17193-2.

- Link (Nov 7, 2011)

Last Updated on Monday, 07 November 2011 13:49

TAUMIE

Mie Type Codes

Written by Pine, D.

Friday, 28 October 2011 07:48

CDA

Multiple Particle Scattering

Written by Baptiste Auguie

Wednesday, 27 July 2011 12:32

Coupled-dipole simulations of sparse plasmonic nanoparticle assemblies using the cda package

Last Updated on Wednesday, 27 July 2011 12:43

Read more...

TMATRIX_D

T-Matrix Codes

Latest Additions

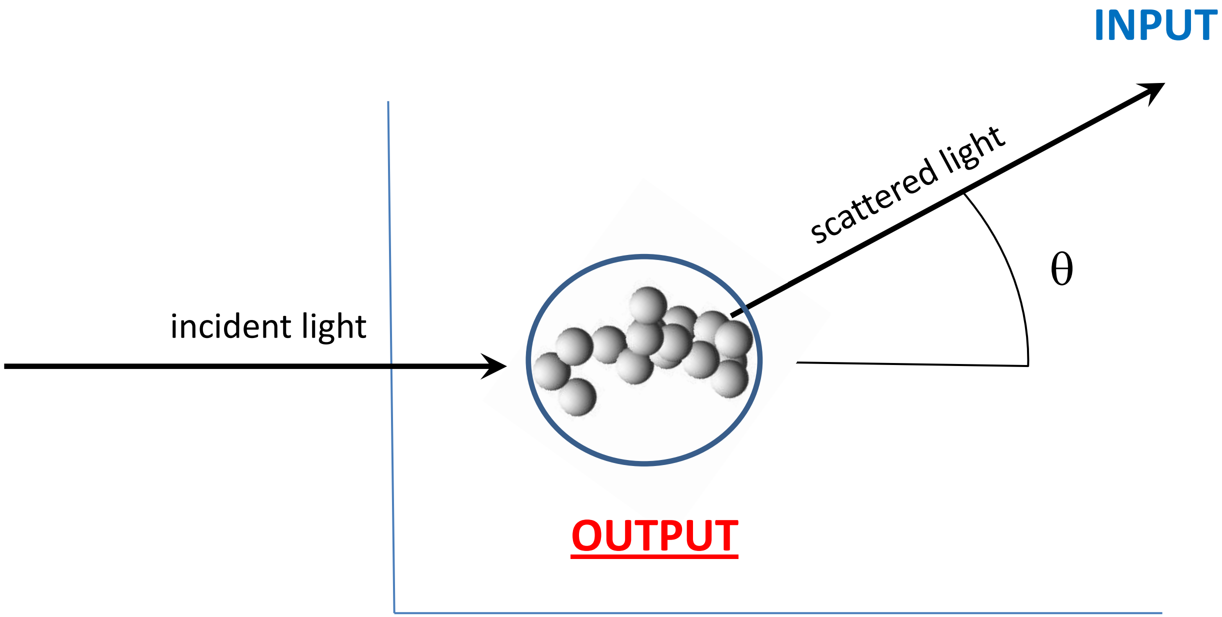
- 4th Conference on Computational Thermal Radiation in Participating Media (CTRPM-4)
- Workshop Scattering 2012
- WORKSHOP INVERSE PROBLEMS 2012
- LIP2012, Rouen 2012
- 3D GLMT
- taumie
- Mélina
- ELS 13, Taormina, 2011
- CDA
- Coupled-dipole simulations of sparse plasmonic nanoparticle assemblies using the cda package

http://www.t-matrix.de/

≈300 free numerical codes of electromagnetic scattering by particles or surfaces!

WHY IS THE INVERSE SCATTERING PROBLEM SO DIFFICULT?

- ***inverse problem*** → we know the Stokes parameters, what is the scatterer?



INPUT = Stokes parameters $S_i(\theta, \lambda)$
OUTPUT = scatterer

the information is spread over 4 functions of <u>a number</u> of variables and parameters

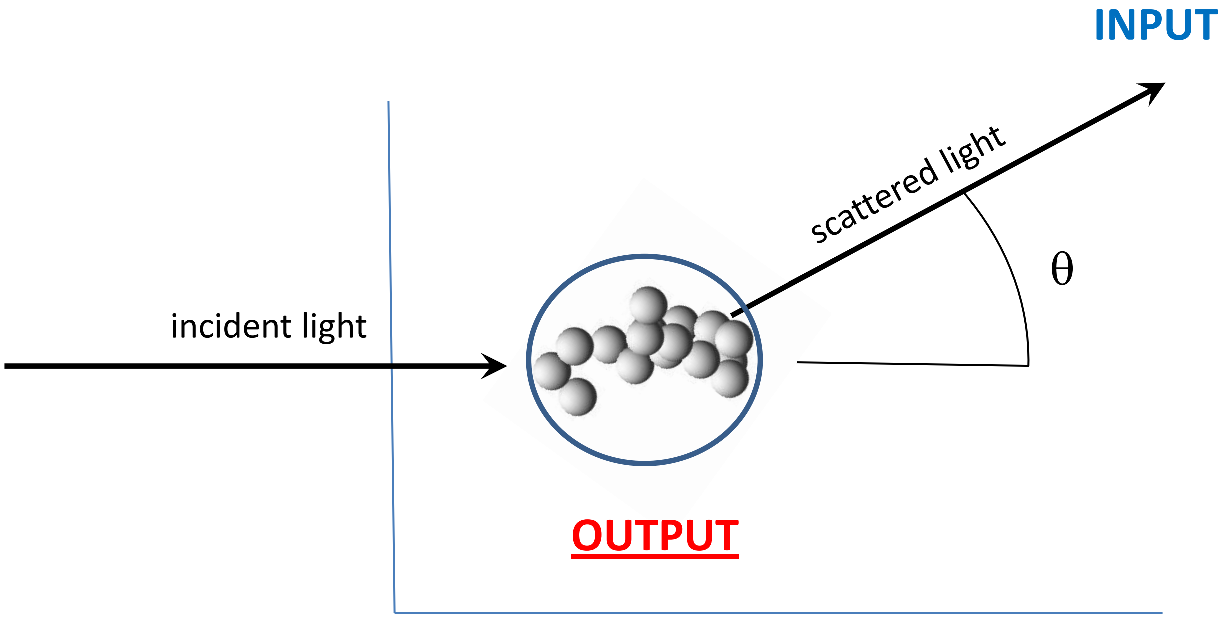
THE FREE NUMERICAL PROGRAMS TO DO SO…

NONE

though all the information exists in the Stokes parameters functions….

WHY IS THE INVERSE SCATTERING PROBLEM SO DIFFICULT?

- ***inverse problem*** → we know the Stokes parameters, what is the scatterer?

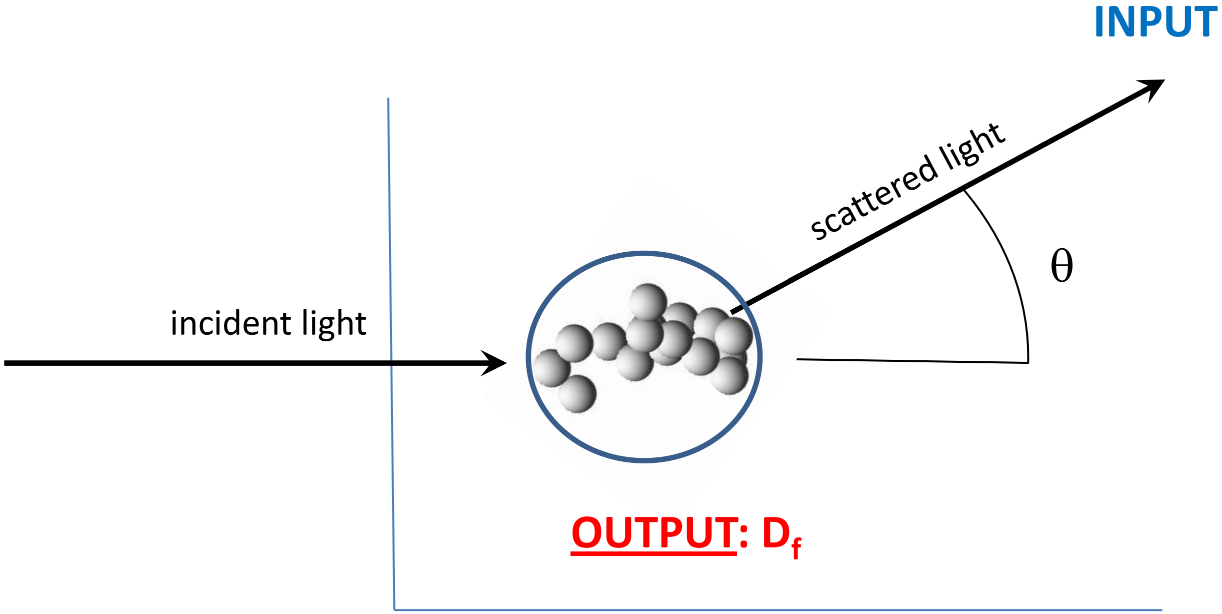


- ***The inverse problem would be simple if we knew where is hidden the value of this or that parameter in the Stokes parameter functions***

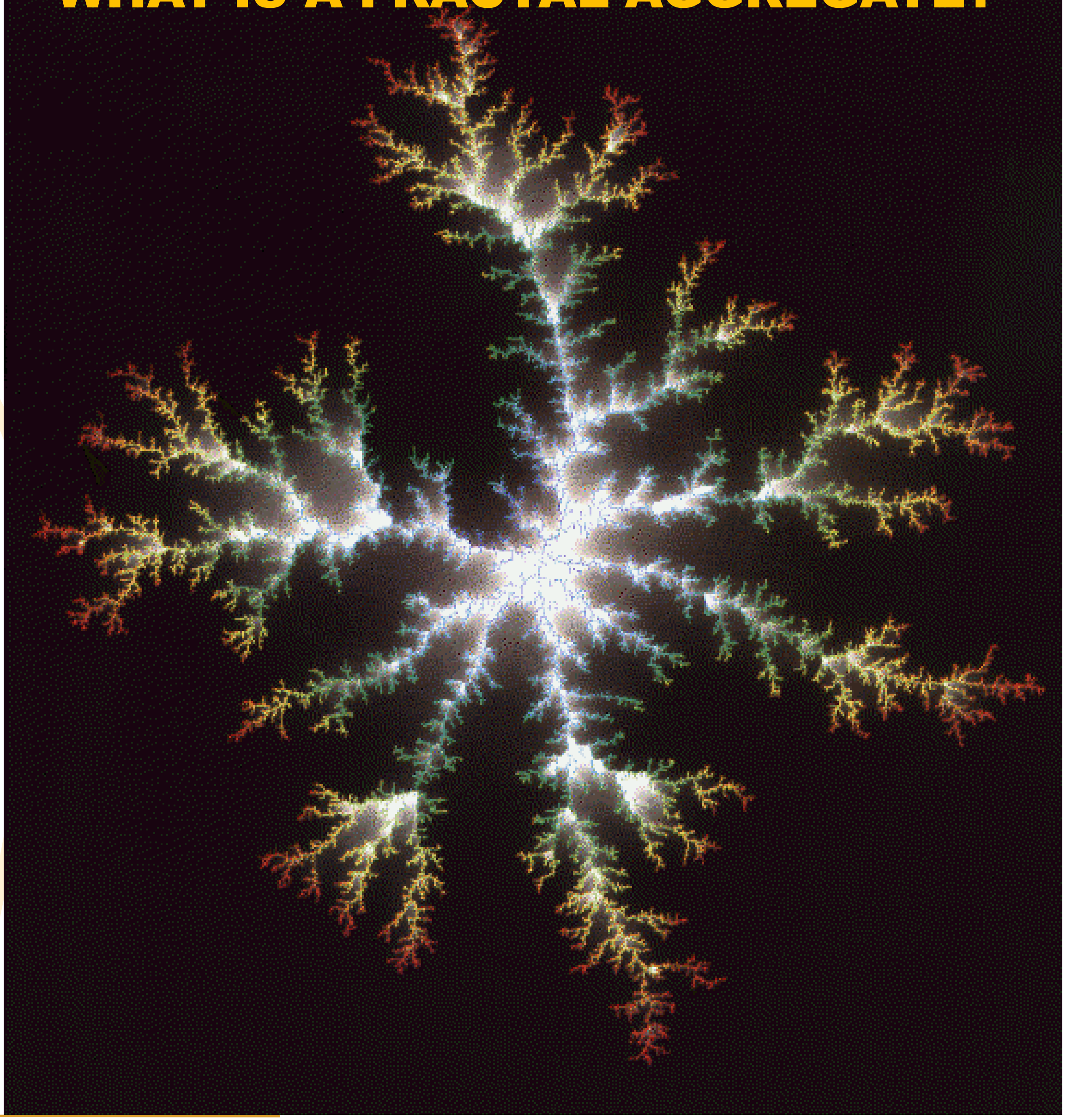
actually, some examples are known but they are rare…

EXAMPLE OF AN INVERSE SCATTERING PROBLEM: / ESTIMATE THE FRACTAL DIMENSION /

- Inverse problem → ***we know the intensity scattered by fractal aggregate of homogeneous grains, what is the value of the fractal dimension***?

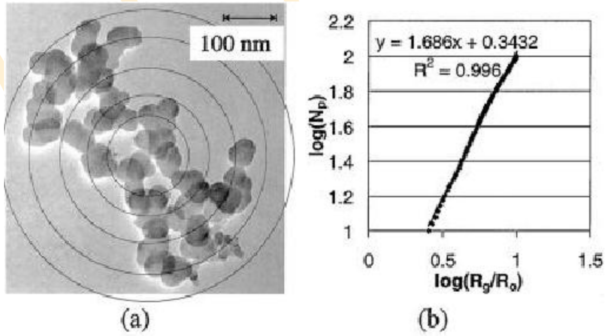


INTERLUDE:
WHAT IS A FRACTAL AGGREGATE?

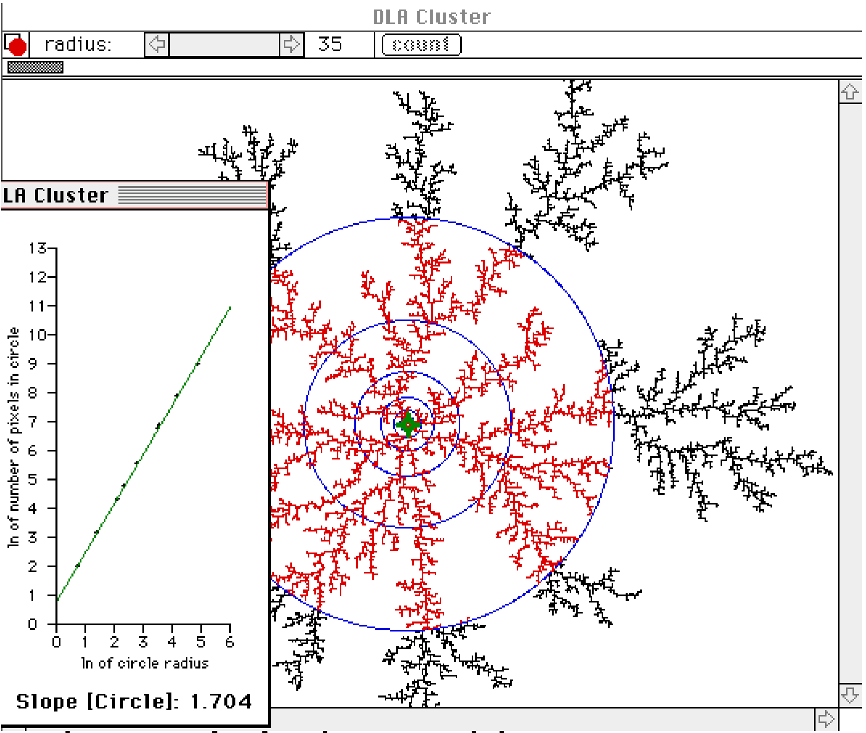


REMINDER OF WHAT IS A FRACTAL AGGREGATE

- In a diluted cold fluid, grains tend to stick together to form fractal aggregates, following a process called: Cluster-Cluster Aggregation (CCA)



(a) (b)



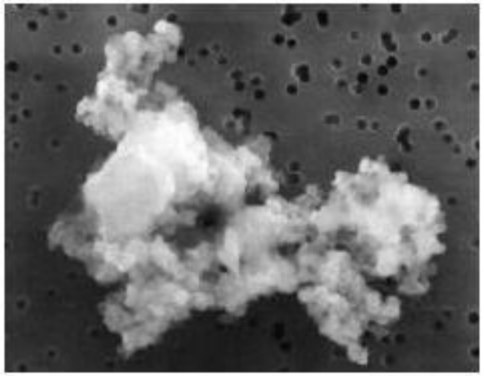
Slope [Circle]: 1.704

$$N \cong c\, R^{D_f}$$

- a fractal aggregate is entirely defined (in the statistical sense) by:
  - its reduced radius of gyration, R_g/a
  - its fractal dimension, D_f
  - Its prefactor, c

FRACTAL AGGREGATE GALLERY 0.

was it fractal before collection?
at least → this is aggregate of grains!



Brownlee particle

FRACTAL AGGREGATE GALLERY 1.

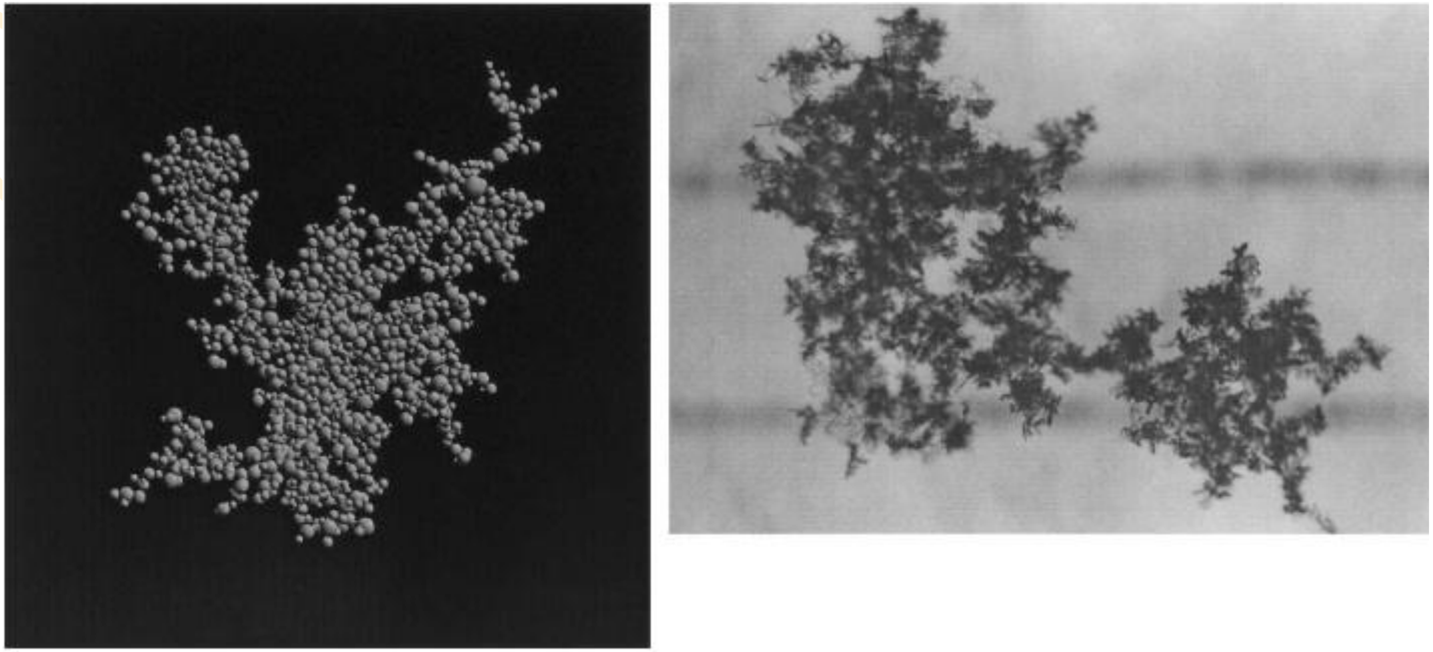
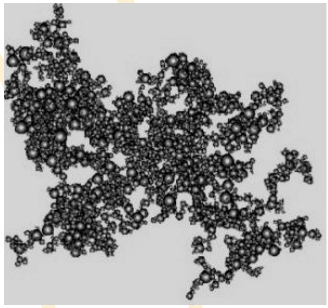


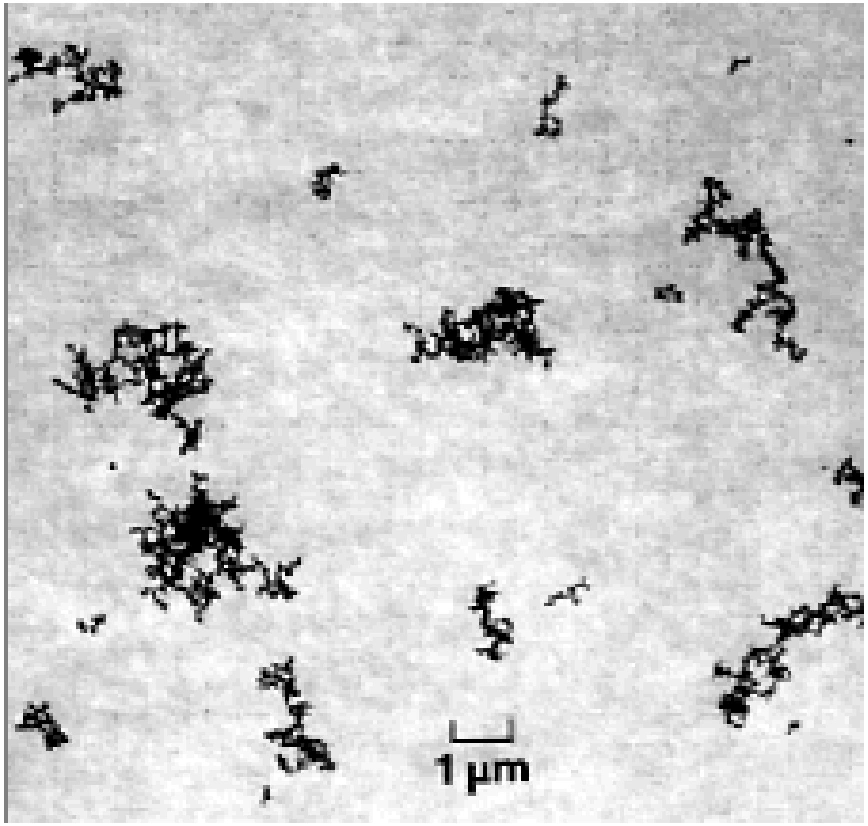
FIG. 2. Images in 3D of a generated snowflake consisting of 1760 particles: (a) top view and (b) front view. (c) A picture of an observed snowflake is also shown for comparison; the two faint lines behind the observed snowflake are separated by 5 mm.

snowflakes (*Maruyama & Fujiyoshi, 2005*)

FRACTAL AGGREGATE GALLERY 2.



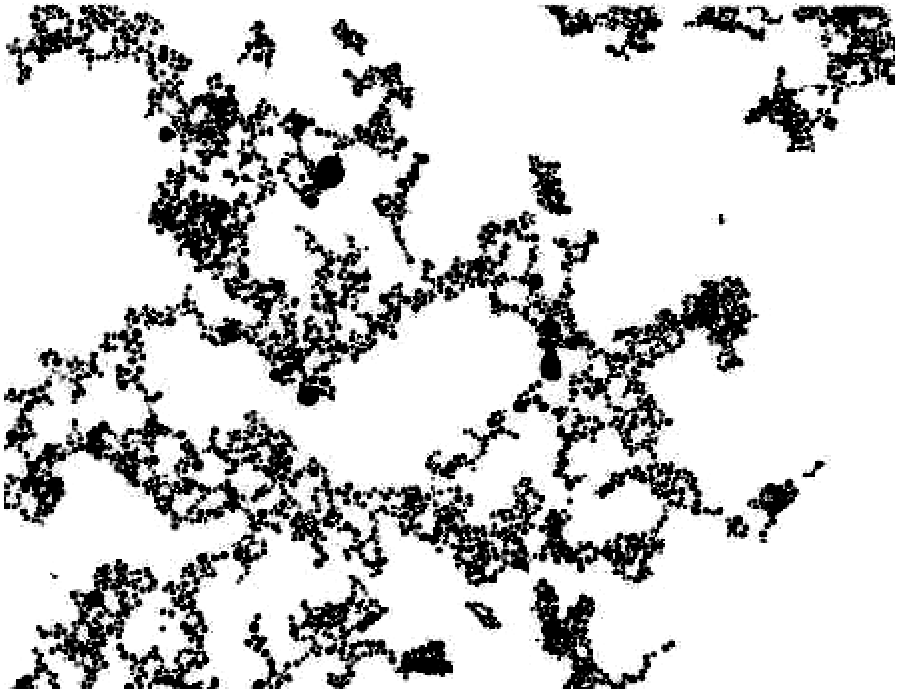
numerical



Au colloidal particles (*Weitz et al, 1985*)

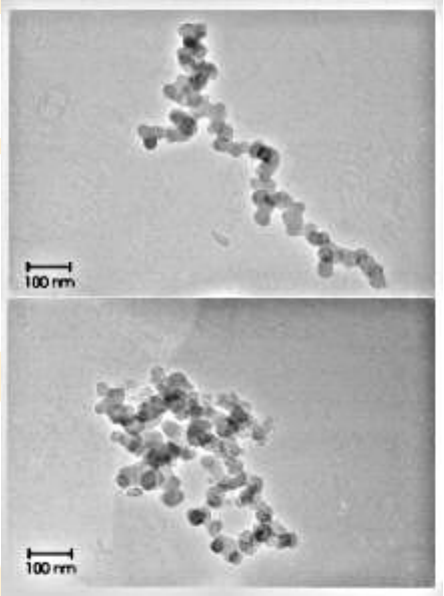
FRACTAL AGGREGATE GALLERY 3.

numerical

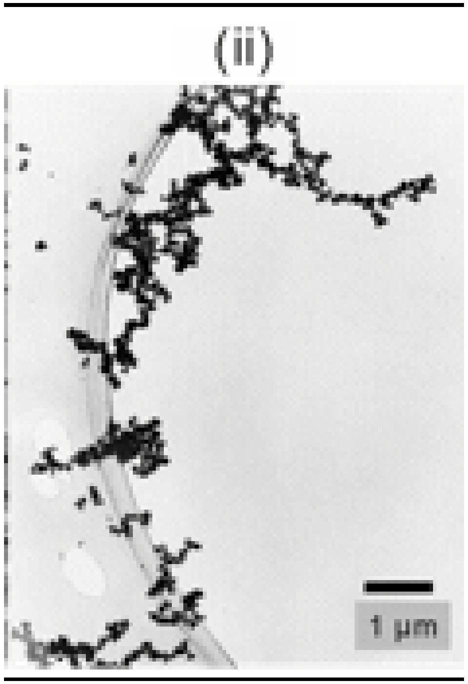


Latex colloidal particles

FRACTAL AGGREGATE GALLERY 4.

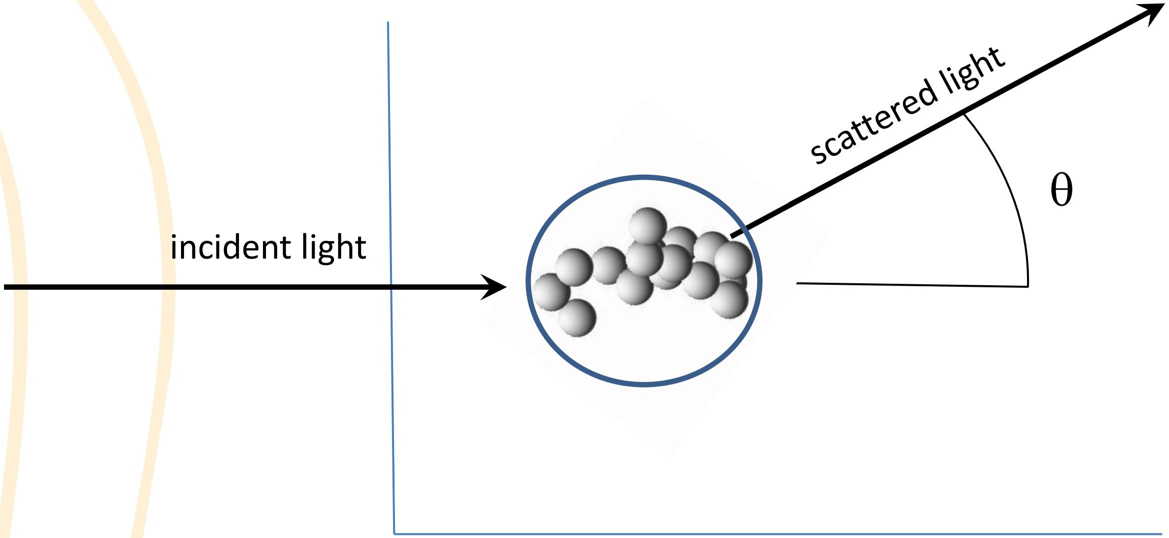


Diesel smoke (*Xiong & Friedlander, 2001*)



laboratory generated meteoric smoke (*Saunders & Plane, 2006*)

THE SAS (SMALL-ANGLE SCATTERING) AS A TOOL TO MEASURE THE FRACTAL DIMENSION OF OPTICAL SOFT PARTICLES



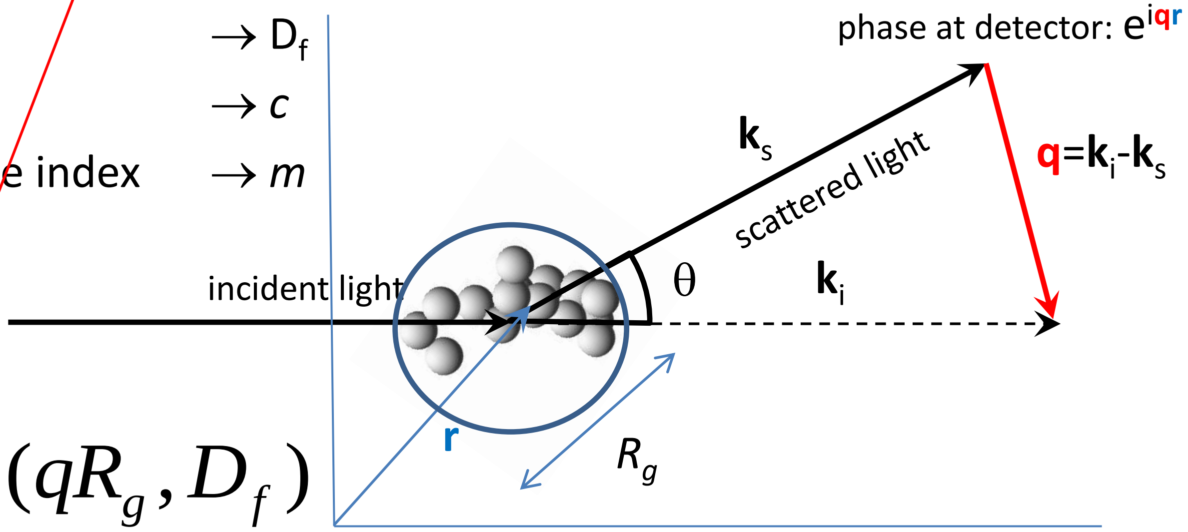
(INTRA) SINGLE-SCATTERING ASSUMPTION…

- Scattering quantities for **isotropic fractal aggregate of homogeneous grains of radius *a* under <u>single-scattering assumption</u>**, must depend on:

5 parameters

- the scattering angle → θ
- the size parameter → $x = 2\pi a/\lambda$
- the shape parameter → R_g/a
- the fractal dimension → D_f
- the prefactor → c
- the complex refractive index → m

single parameter: $qa = 2x\sin(\theta/2)$



$$I_N(q) = c\, I_1(qa, m)\, S(qR_g, D_f)$$

1-grain scattering *structure factor*

$$q = |\mathbf{q}| = \frac{4\pi}{\lambda}\sin(\theta/2)$$

WHAT ABOUT THE STRUCTURE FACTOR?

$$I_N(q) = cI_1(qa,m)\, S(qR_g, D_f)$$

1-grain scattering *structure factor*

depends only on R_g

$$S = \frac{1}{N^2}\left|\sum_j e^{i\mathbf{q}\mathbf{r}_j}\right|^2$$

= exp($-q^2R_g{}^2/3$) when $qR_g < 1$

$$= \frac{A}{\left(qR_g\right)^{D_f}}$$ when $1 < qR_g$

scaling with exponent D_f

$$q = |\mathbf{q}| = \frac{4\pi}{\lambda}\sin(\theta/2)$$

PUTTING ALL TOGETHER (SAS = VERSUS θ)

In terms of the quantities θ et λ, these results write:

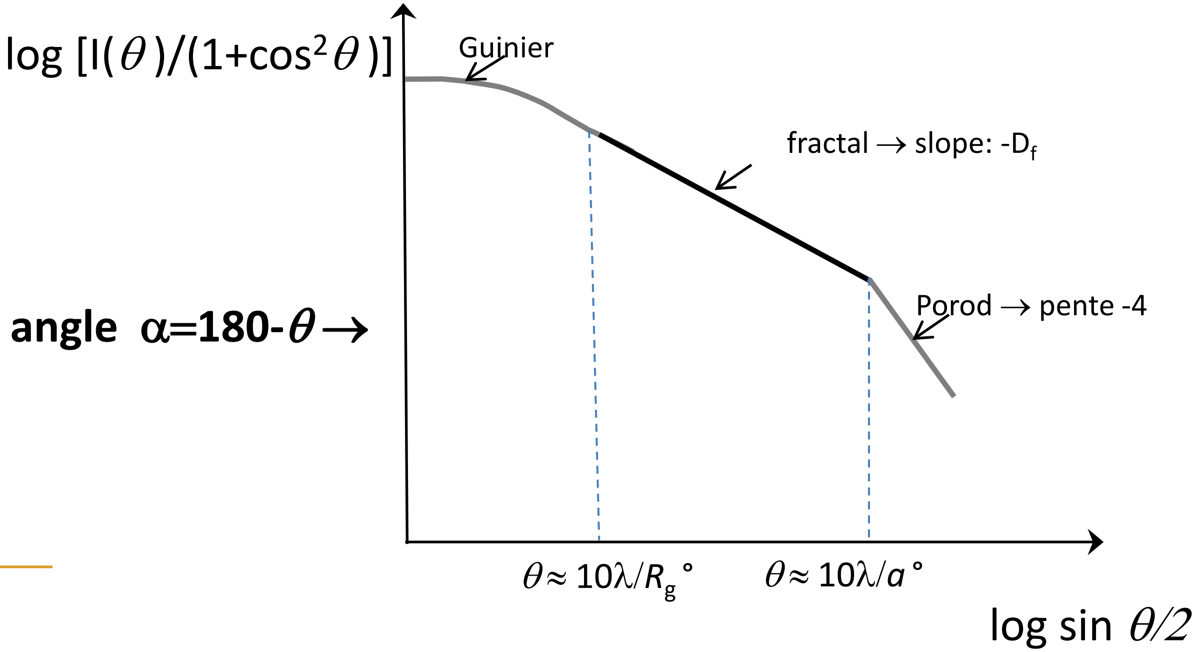
Rayleigh $\propto (1+\cos^2\theta)/\lambda^4$

Structure factor → S = total intensity/intensity scattered by a grain

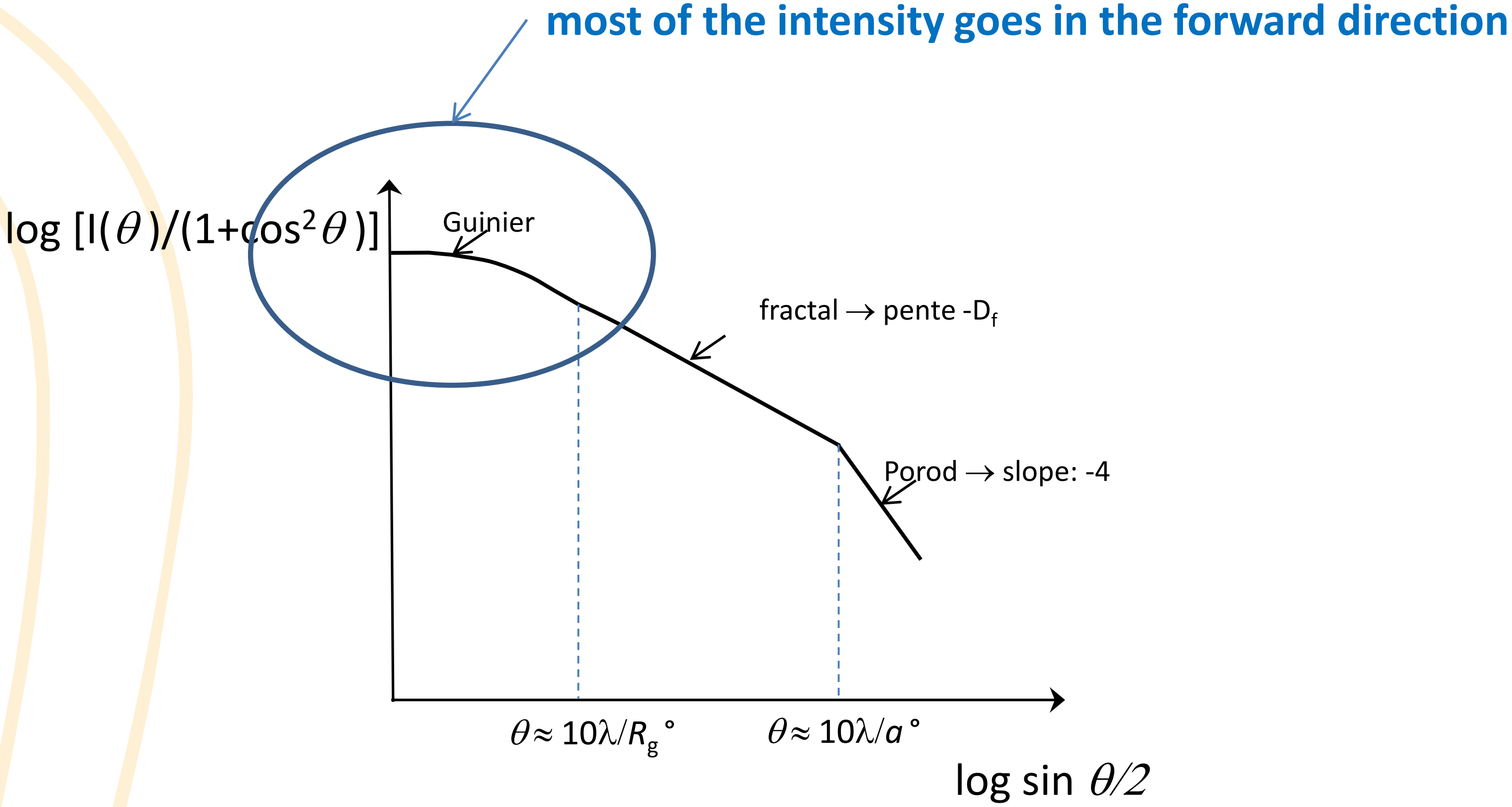
$S \sim \exp(-c(\theta R_g/\lambda)^2)$ for the small values of θ (Guinier regime)

$\sim (\lambda/\sin(\theta/2))^{D_f}$ for the intermediate values of θ (fractal regime)

varying the phase angle α=180-θ →

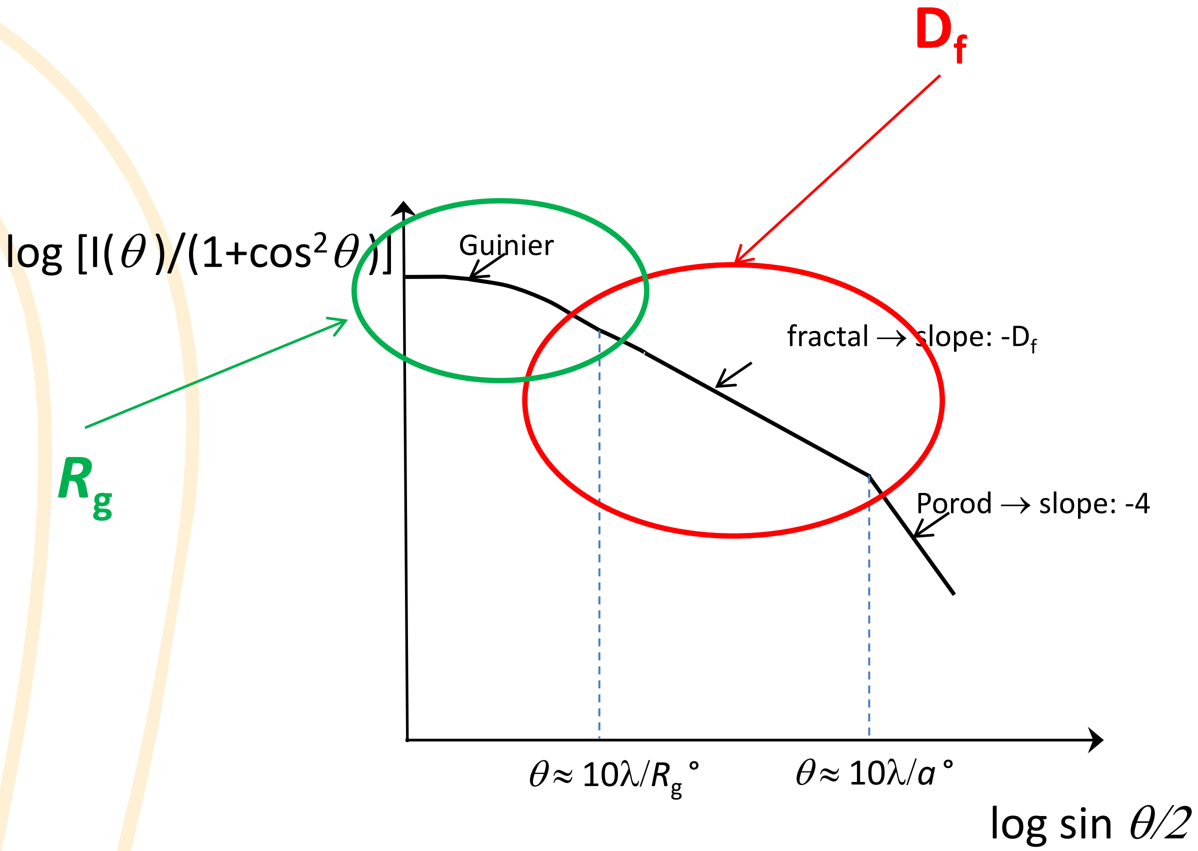


PUTTING ALL TOGETHER (SAS = VERSUS θ)



- **Conditions**:
  - **|m-1|<<1** (optically soft particles)
  - **|2ka(m-1)|<<1** (small grains)

2 PARAMETERS CAN BE MEASURED IN SAS : R_g AND D_f



- **Conditions**:
  - $|m-1| \ll 1$ (optically soft particles)
  - $|2ka(m-1)| \ll 1$ (small grains)

PUTTING ALL TOGETHER (VERSUS λ)

In terms of the quantities θ et λ, these results write:

Rayleigh $\propto (1+\cos^2\theta)/\lambda^4$

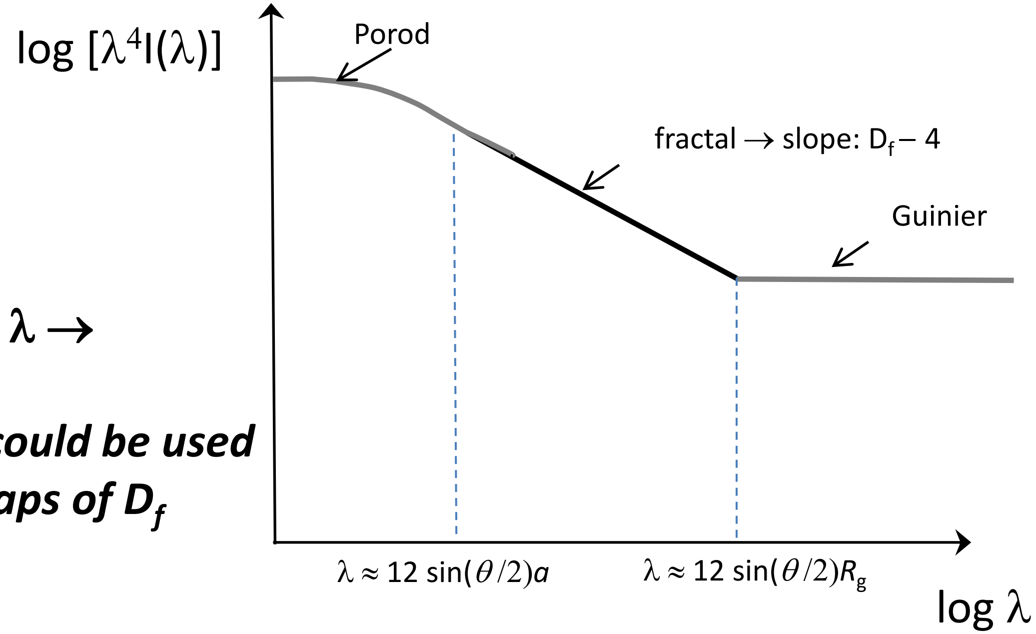
Structure factor → S = total intensity/intensity scattered by a grain

$S \sim \exp(-c(\theta R_g/\lambda)^2)$ for the large values of λ

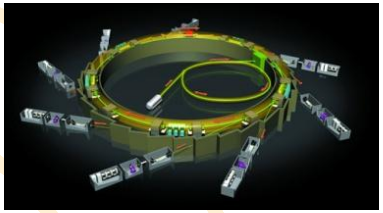
$\sim (\lambda/\sin(\theta/2))^{D_f}$ for the intemediate values of λ

varying the wavelength λ →

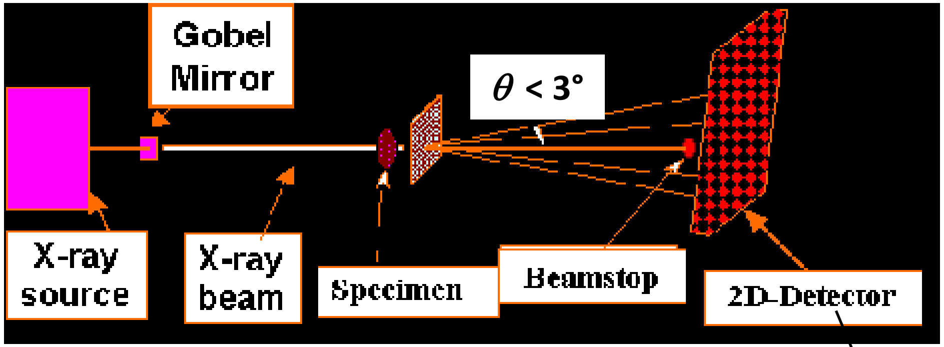
ultraspectral imaging could be used to have spatial maps of D_f

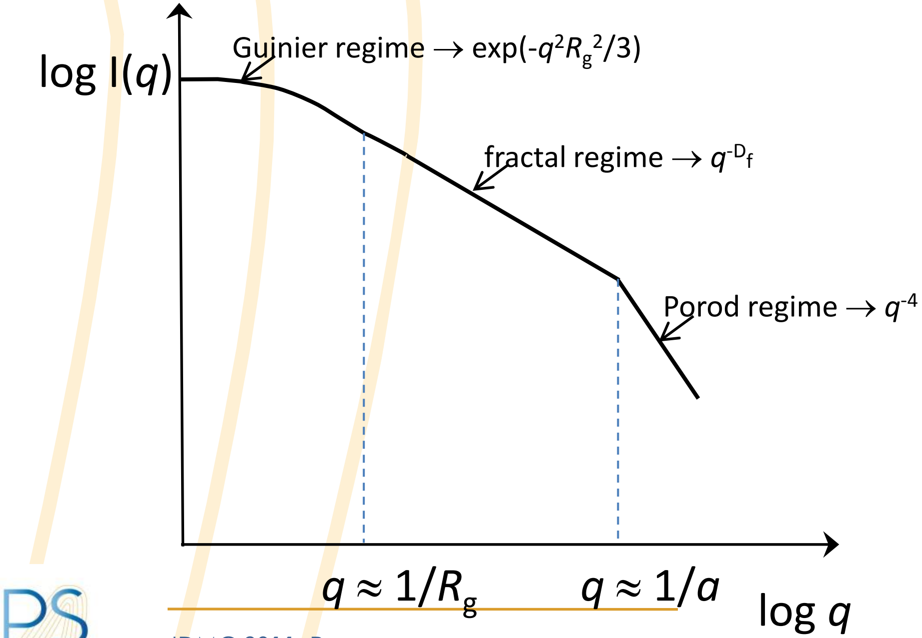


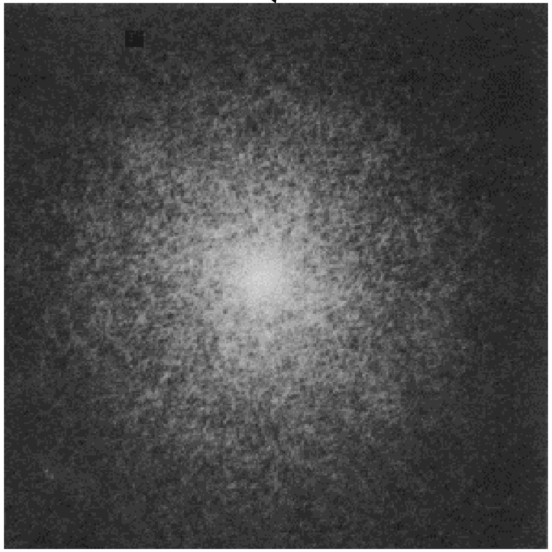
CURRENT APPLICATION : SAXS



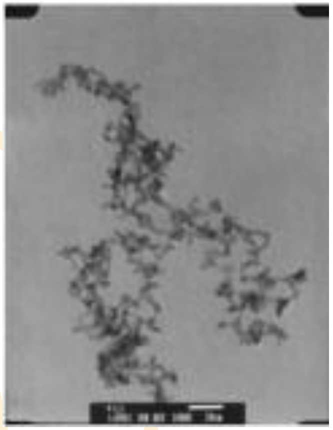
Soleil synchroton



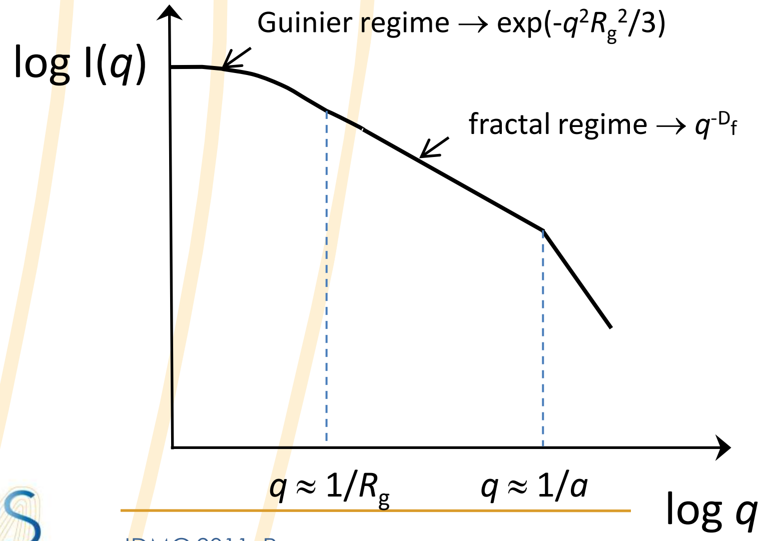




CURRENT APPLICATION : SAXS



Pt fractal nanoparticles (*Min et al, 2007*)



log I(q)

Guinier regime → $\exp(-q^2 R_g^2/3)$

fractal regime → q^{-D_f}

$q \approx 1/R_g$

$q \approx 1/a$

log q

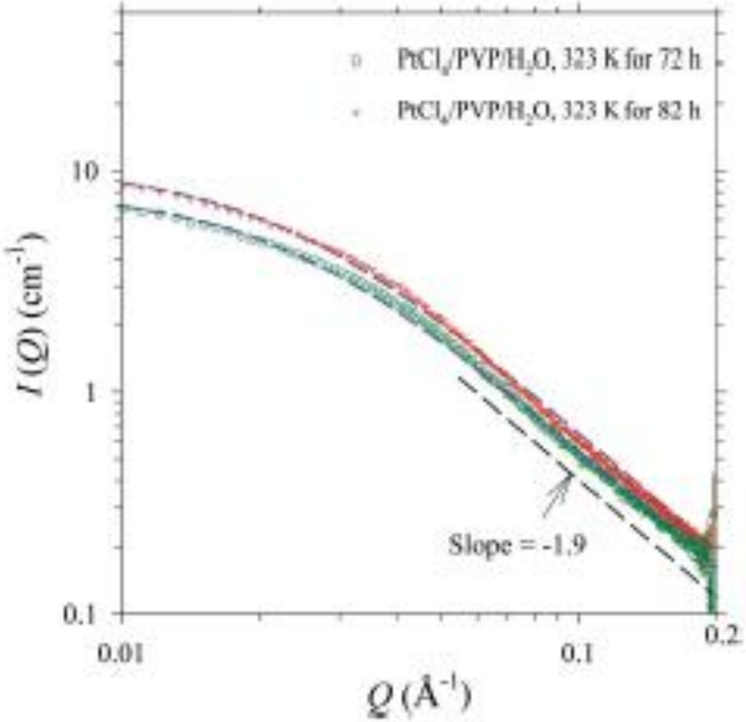
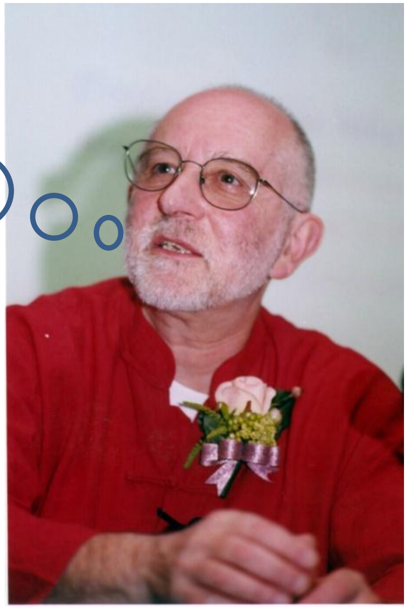


Figure 4
SAXS data for the aqueous solution of PtCl$_4$/PVP after 72 and 82 h annealing at 323 K.

…BUT WHAT ABOUT MULTIPLE-SCATTERING???

multiple-scattering is irrelevant for $D_f \leq 2$, in the mean-field approximation

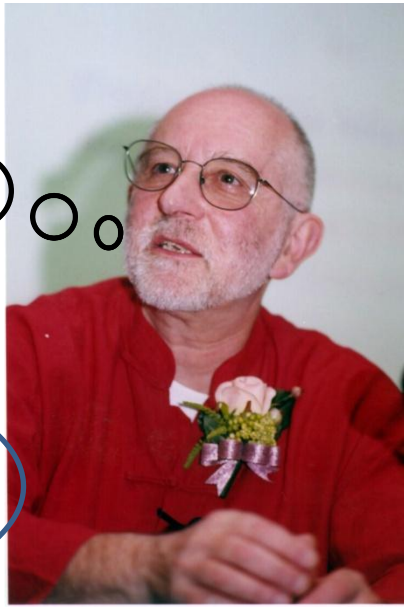
(*i.e. the em fields are similar inside each grain*)



Berry & Percival, 1986

…BUT WHAT ABOUT MULTIPLE-SCATTERING???

multiple-scattering is irrelevant for $D_f \leq 2$,
in the mean-field approximation
(*i.e. the em fields are similar inside each grain*)



Berry & Percival, 1986

Berry is correct!

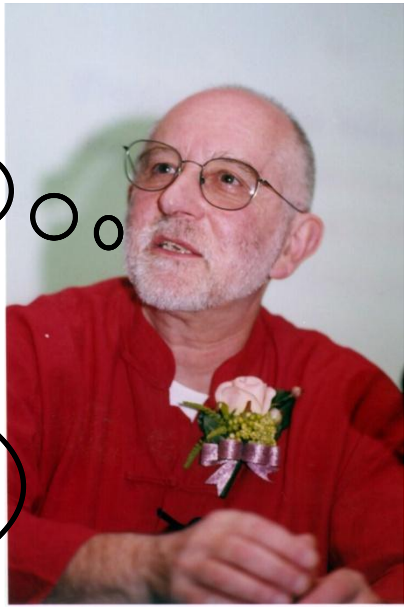
$$\left.\begin{array}{ll} \mathbf{I(\mathit{N})/I(1) \sim q^{-D_f}} & \textbf{for } \mathbf{D_f \leq 2} \\ \mathbf{\sim q^{-2}} & \textbf{for } \mathbf{D_f>2} \end{array}\right\} 1/R_g << q << 1/a$$

within the mean-field assumption

Botet et al, 1995

...BUT WHAT ABOUT MULTIPLE-SCATTERING???

multiple-scattering is irrelevant for $D_f<2$, in the mean-field approximation (*i.e. the em fields are similar inside each grain*)



Berry & Percival, 1986

Botet et al, 1995

Berry is correct!

$$\mathbf{I(\mathit{N})/I(1)} \sim \left.\begin{array}{ll} \mathbf{q^{-D_f}} & \mathbf{for\ D_f \leq 2} \\ \mathbf{q^{-2}} & \mathbf{for\ D_f>2} \end{array}\right\} \ 1/R_g << q << 1/a$$

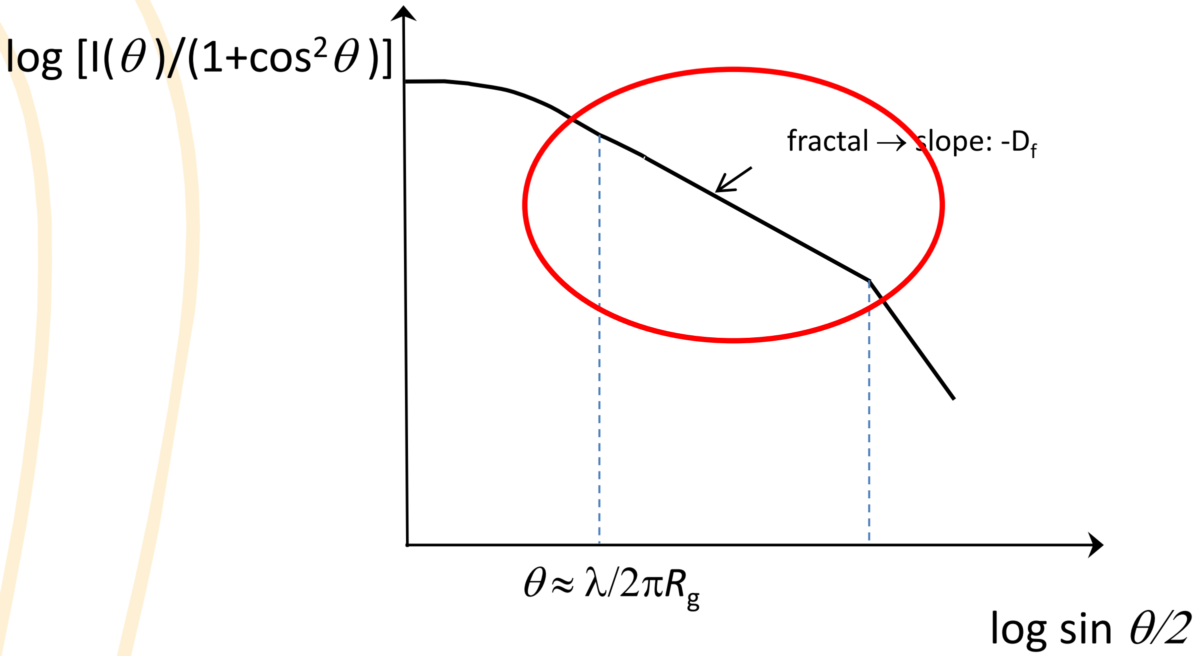
within the mean-field assumption

the mean-field assumption is valid when the material is far from optical resonance

Shalaev, 2000

…AND THE CONCLUSION ABOUT MULTIPLE-SCATTERING

The fractal relation $S(q) \sim q^{-D_f}$ is robust!
It is valid whenever $D_f \leq 2$ and material far from resonance



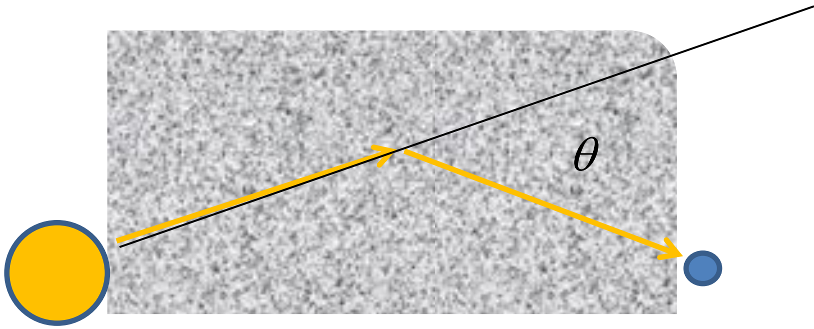
when $D_f > 2$, we lose the information about the fractal dimension because the light is 'trapped' inside loops of particles

– **Conditions**:
  - **off-resonance and $D_f \leq 2$**
  - **ka<<1** (small grains)

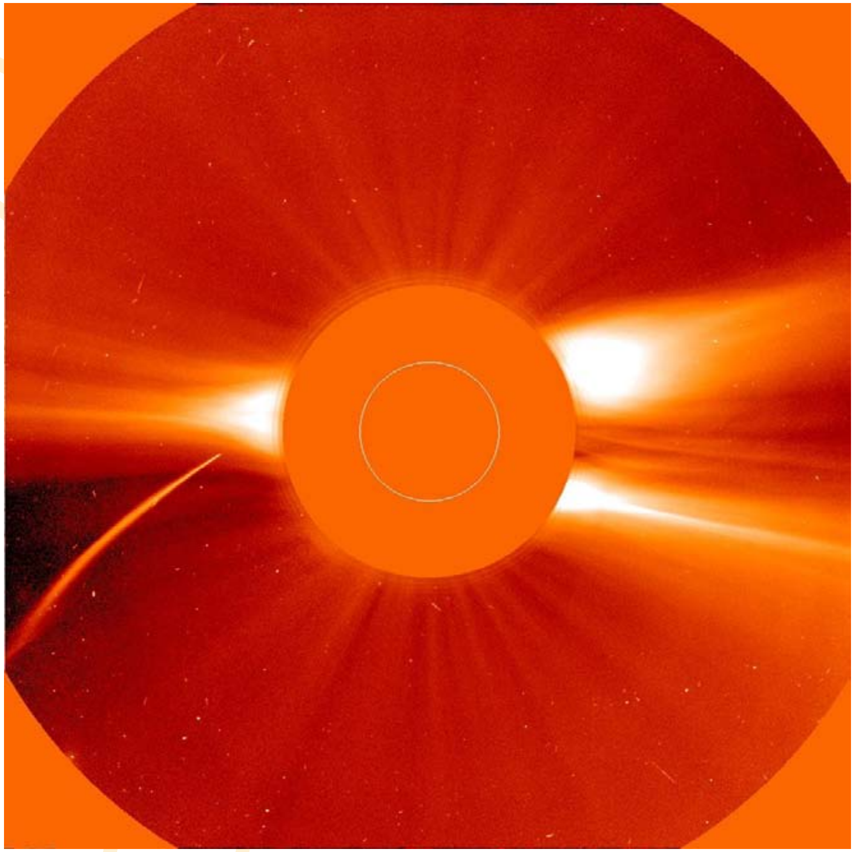
POSSIBLE APPLICATIONS

zodiacal light

(photo: Beletsky)

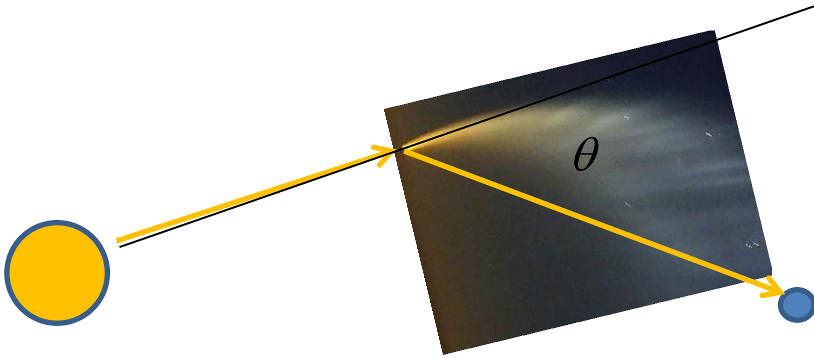


POSSIBLE APPLICATIONS

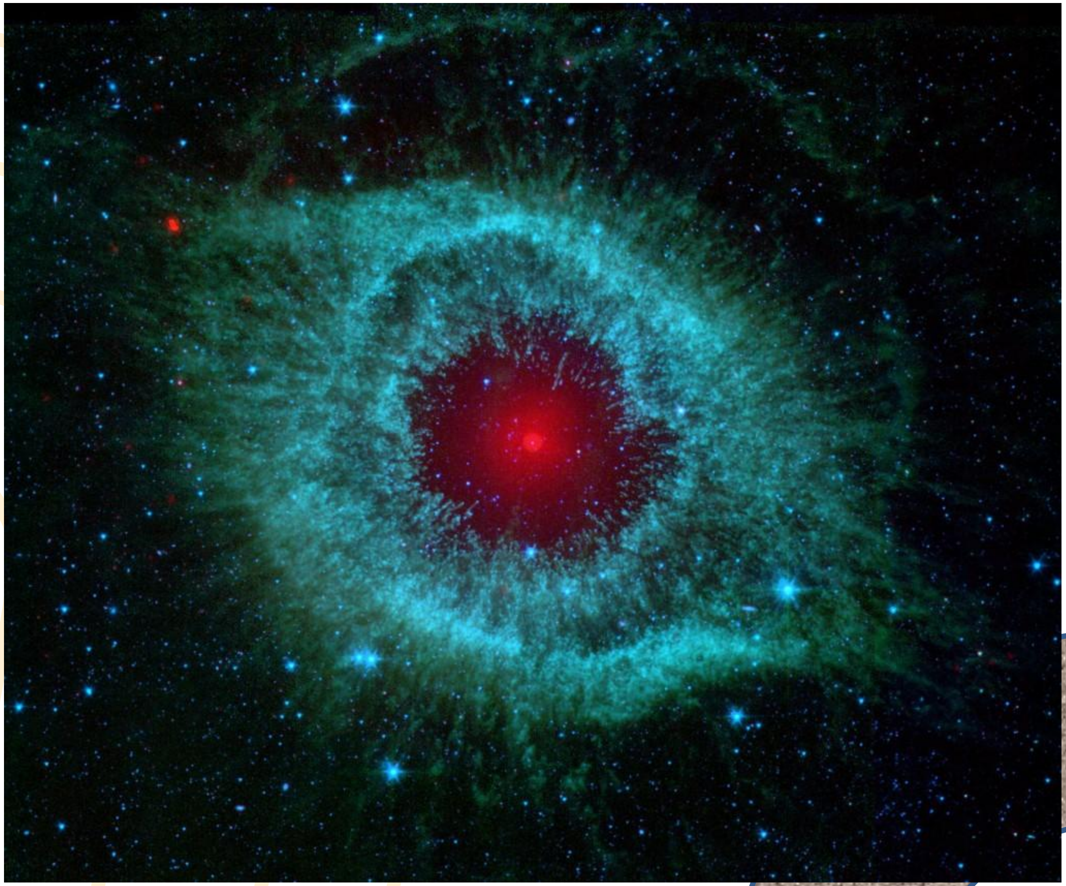


sungrazer comets

(photo: SOHO Consortium)

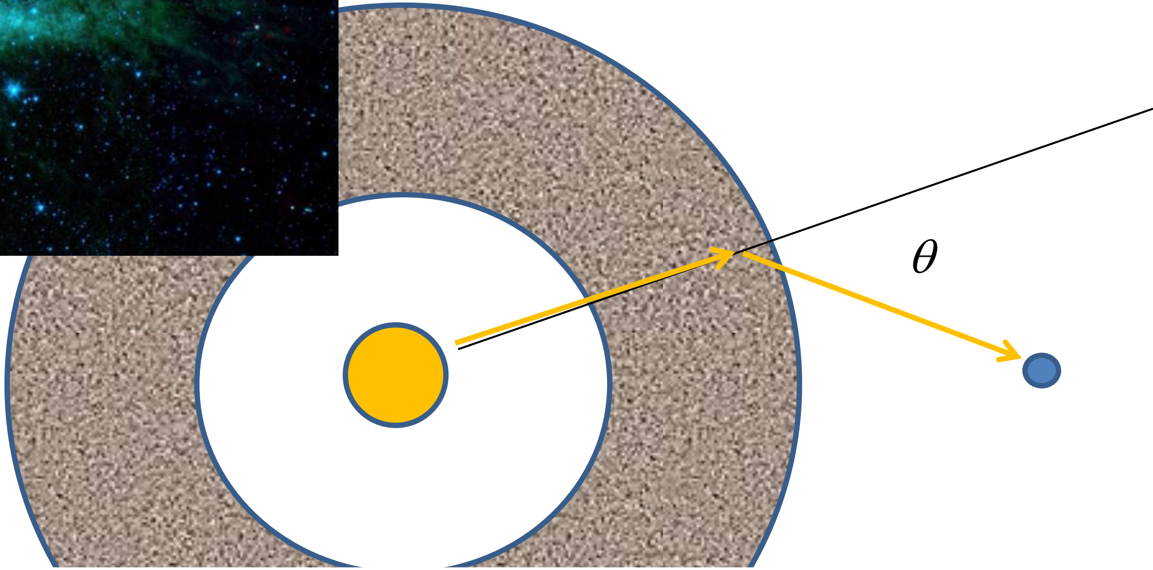


POSSIBLE APPLICATIONS

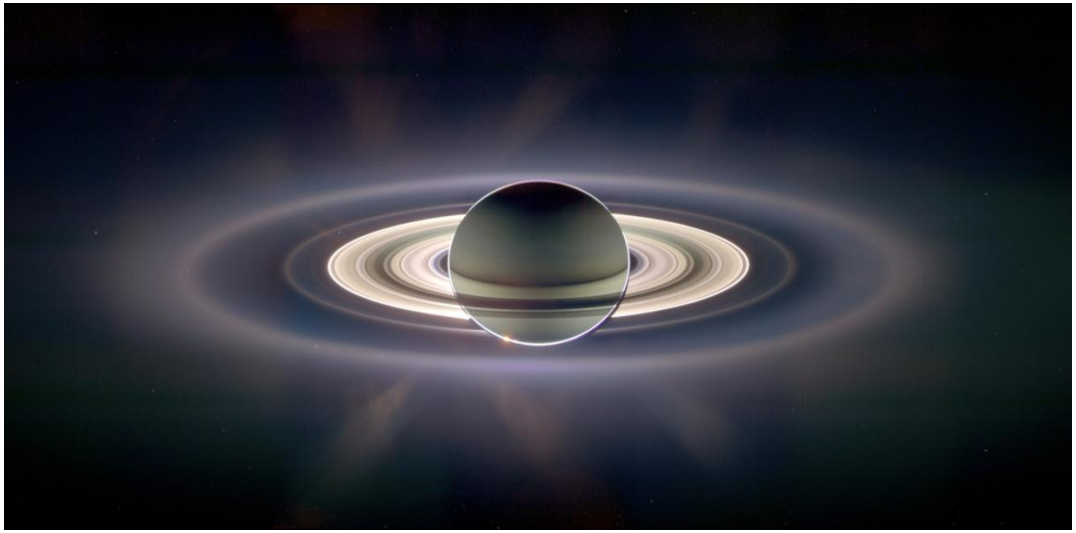


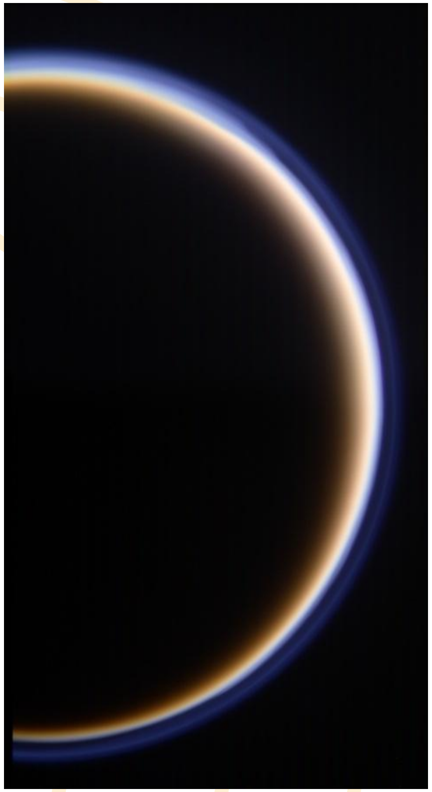
planetary nebula – *infrared image*

*(*photo: NASA's Spitzer Space Telescope)

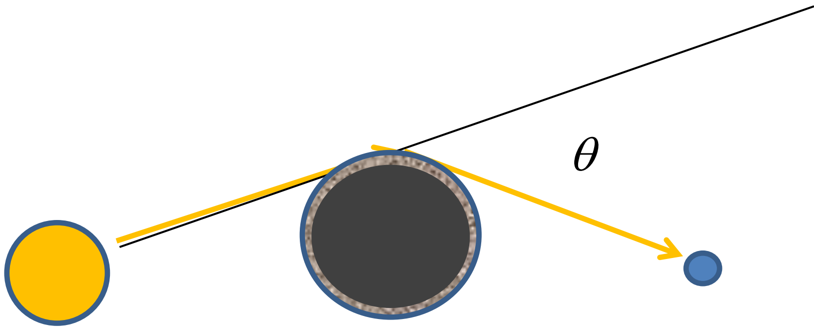


POSSIBLE APPLICATIONS

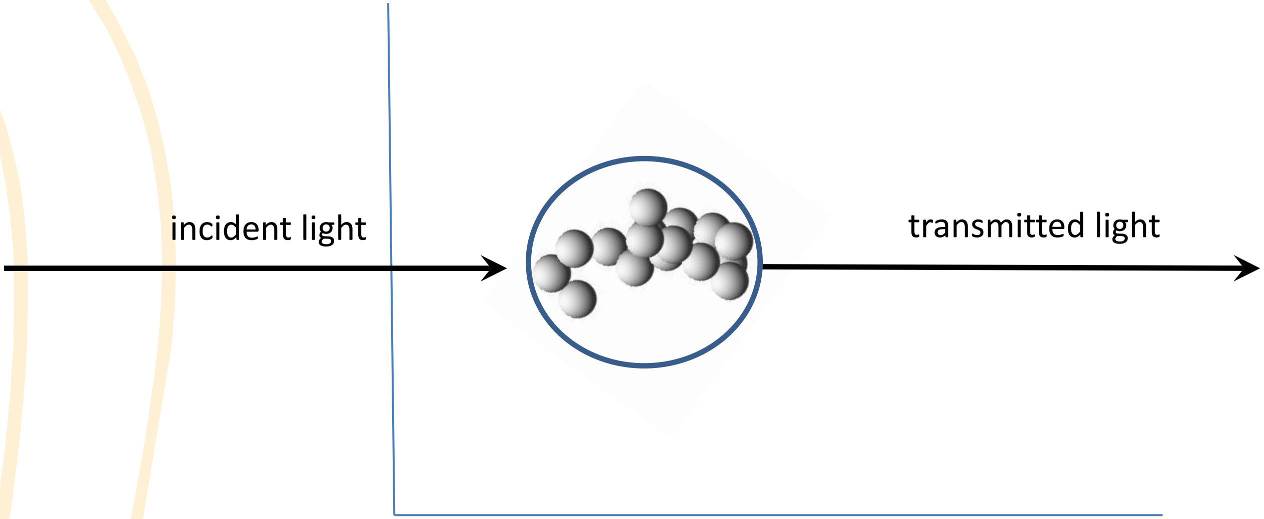




upper atmospheres in forward diffusion
(photos: NASA-JPL)

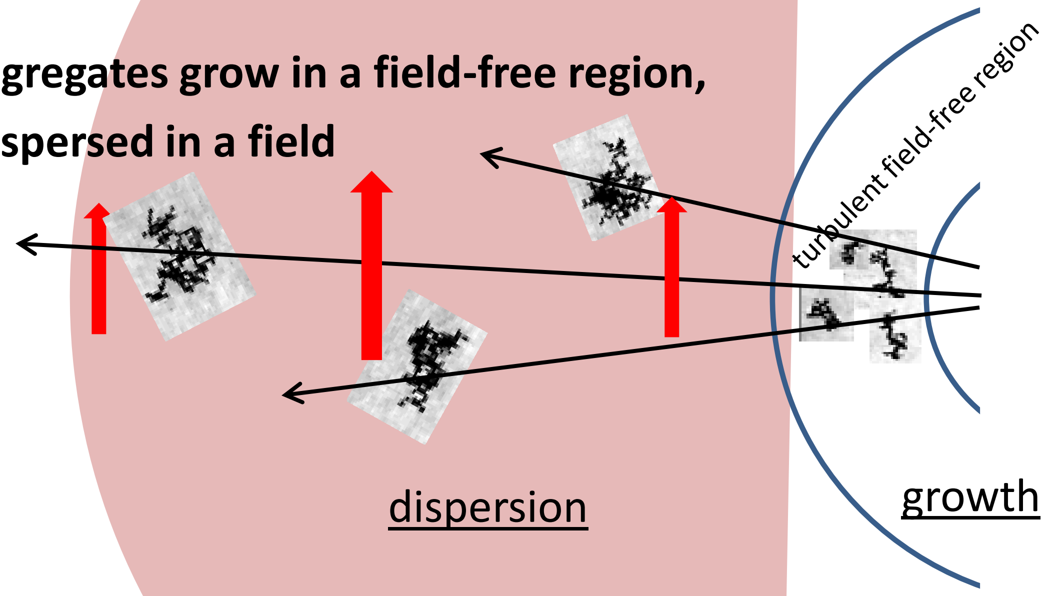


THE POLARIZATION OF FORWARD TRANSMITTED LIGHT AS A TOOL TO MEASURE SIZE OF ALIGNED ABSORBING AGGREGATES



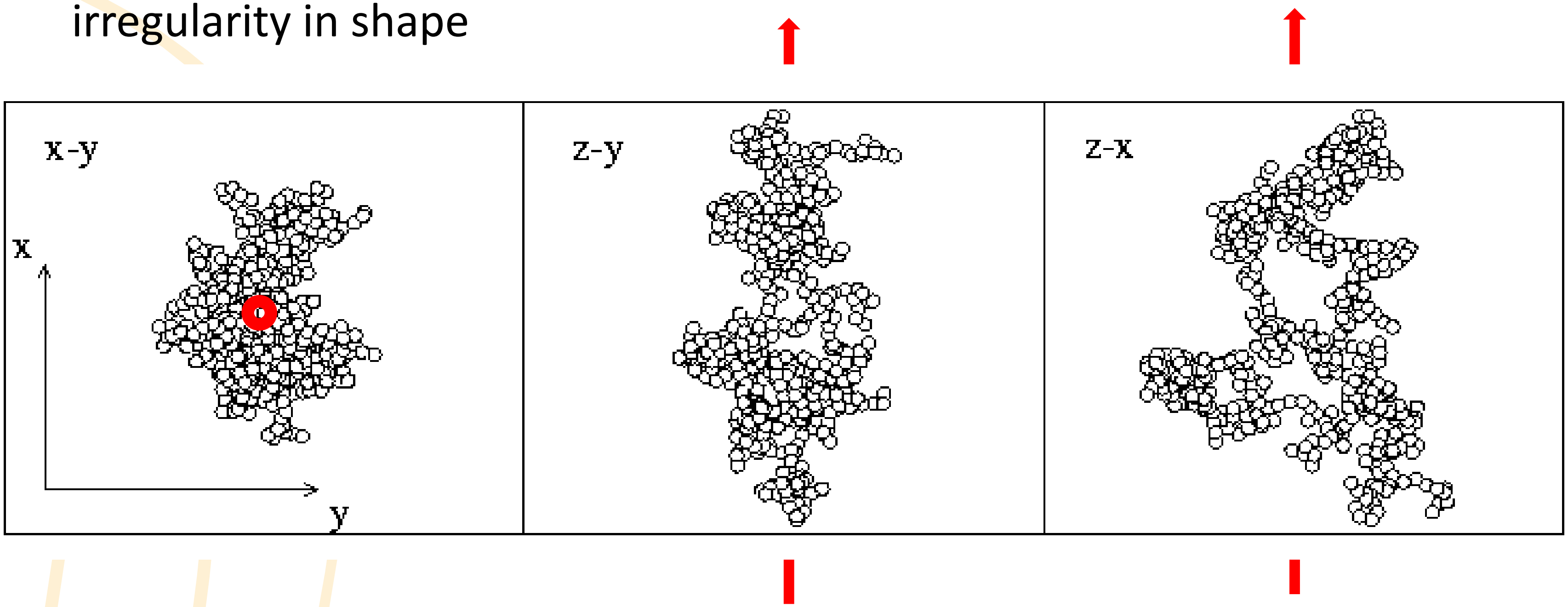
1) FRACTAL AGGREGATES DISPERSED IN A FIELD

- anisotropy of aggregates can appear in 2 different conditions:
  - **1) fractal aggregates grow in a field-free region, then, are dispersed in a field**



THE NATURAL ANISOTROPY OF DISORDERED FRACTAL AGGREGATES

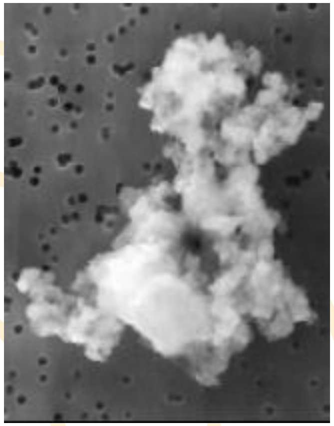
- disordered fractal aggregates are ***statistically*** isotropic
- but when placed in an oriented field, they can exhibit anisotropy due to irregularity in shape



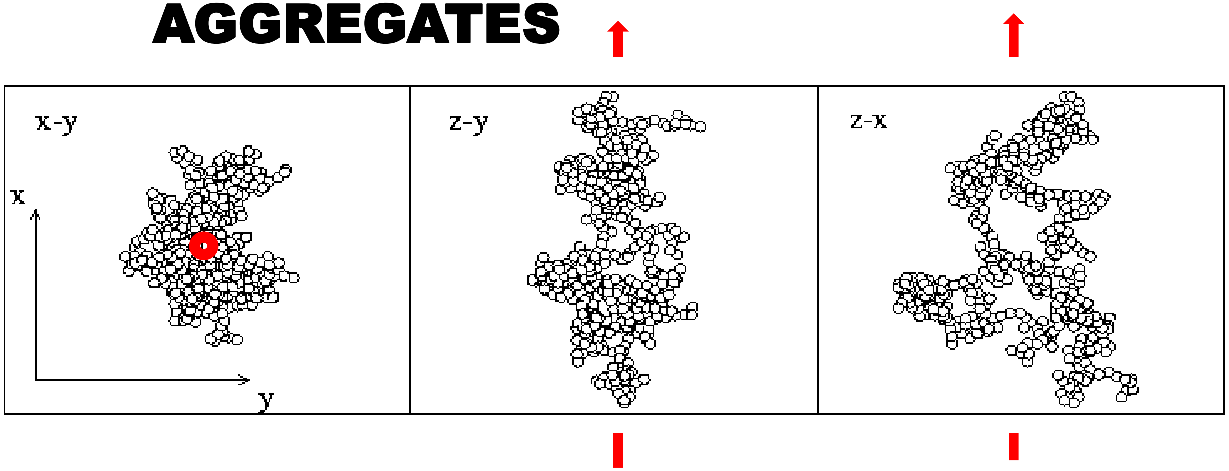
a same N=512 *CCA aggregate of fractal dimension 2, with the axis of smallest inertia moment aligned along z*

- **what about the polarization of the light transmitted through a cloud of such aligned aggregates?...**

THE NATURAL ANISOTROPY OF DISORDERED FRACTAL AGGREGATES

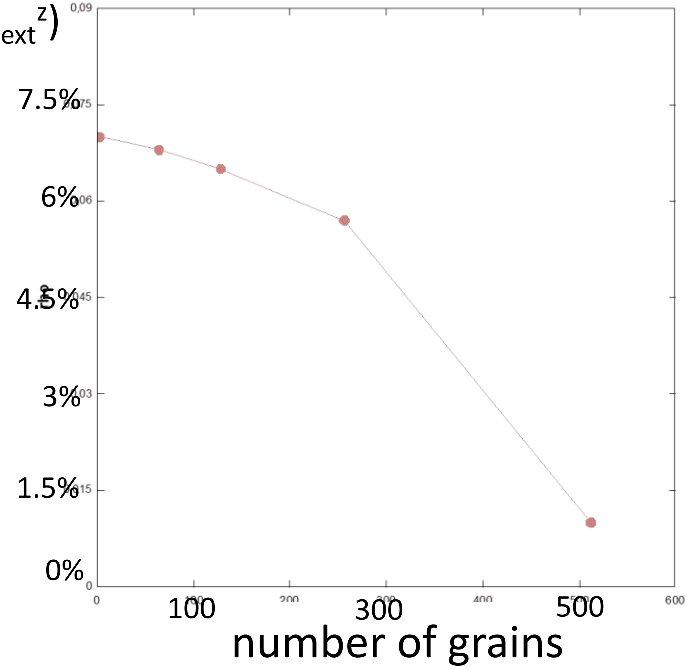


IDP collected in the stratosphere



differential extinction parameter

$\rho = 2(C_{ext}^{y} - C_{ext}^{z})/(C_{ext}^{y} + C_{ext}^{z})$

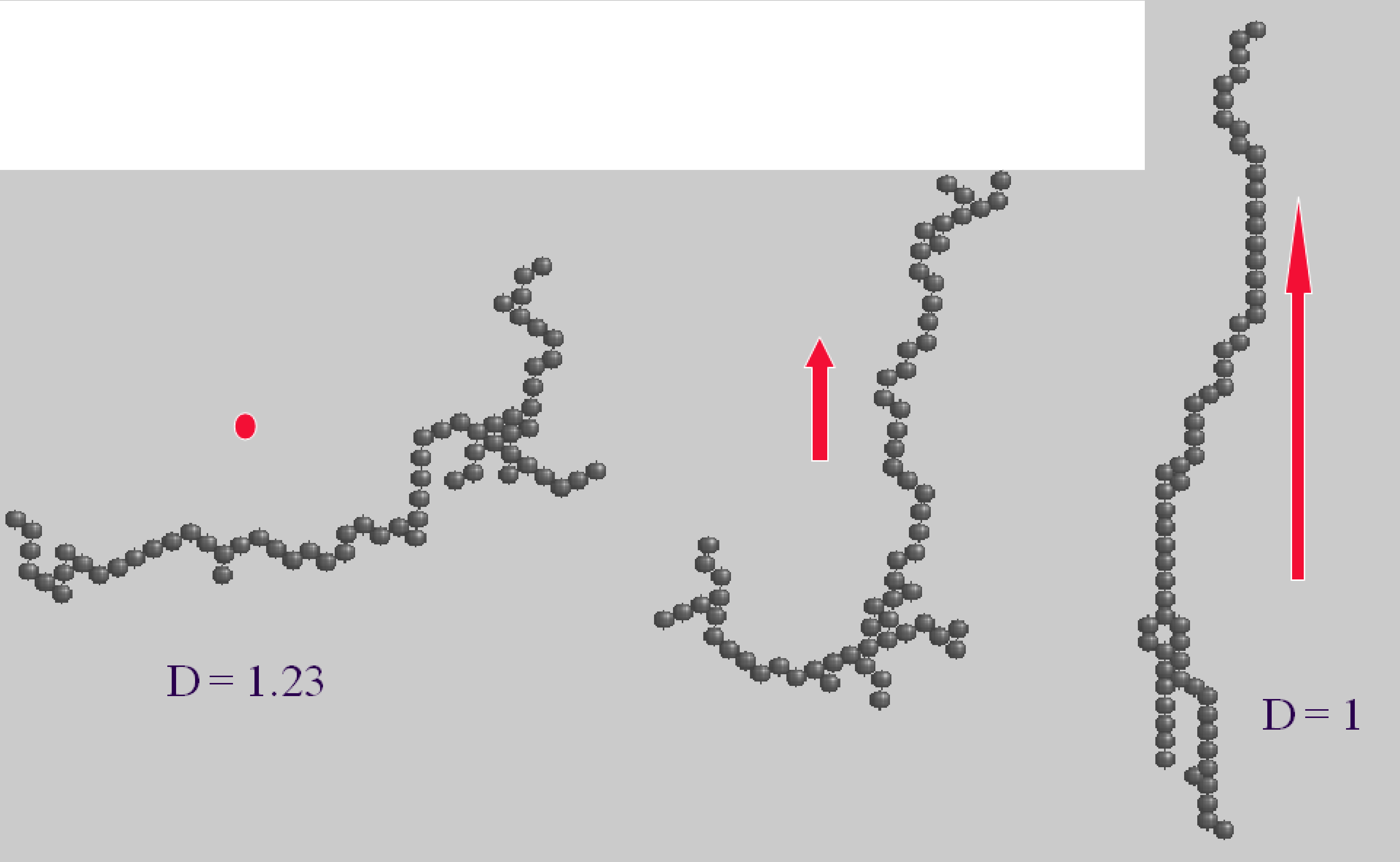


- the polarization due to differential cross-sections decreases with the aggregate size

- **This is an example of relation: polarization degree / size of the particles**

CCA, $D_f=2$, $m=1.4+i\ 0.5$, $2\pi a/\lambda=0.1$, DDA numerical code

2) FRACTAL AGGREGATES GROW IN A FIELD REGION



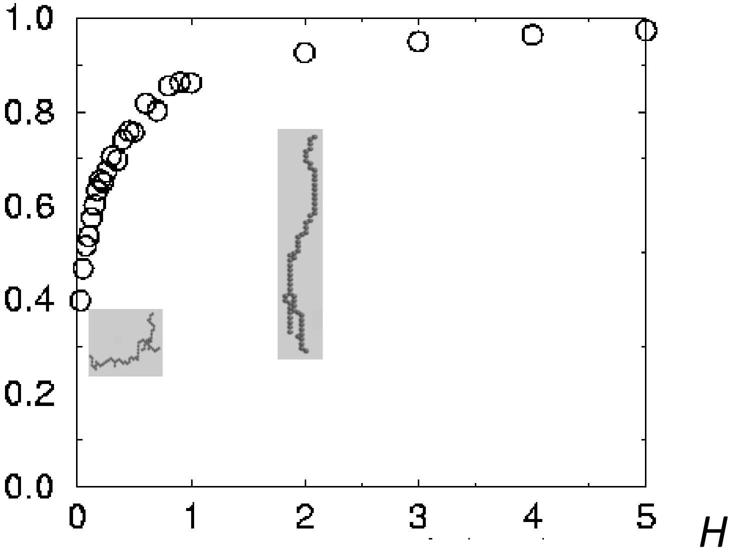
2) FRACTAL AGGREGATES GROW IN A FIELD REGION

- **This case is analogous to a critical system in a field, then:**

$$\textbf{anisotropy} \sim H^{1/\delta}$$

(not so) large fields $\Rightarrow$ aggregates $\approx$ rods

(…but precise studies are lacking…)



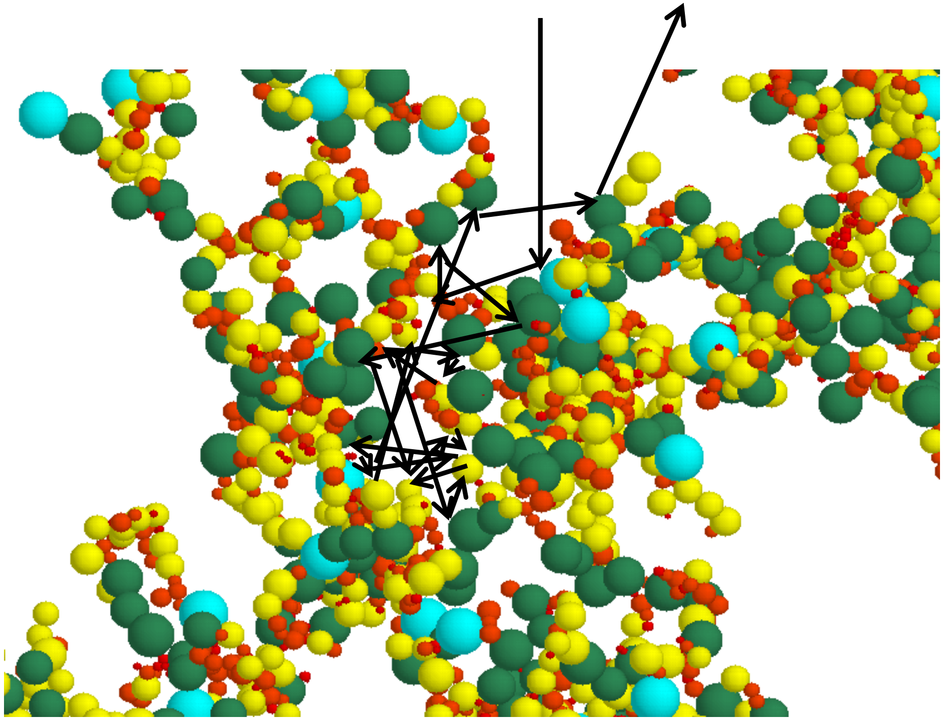
ANOMALOUS AD/ABSORPTION ON FRACTAL AGGREGATES
AND CONSEQUENCES

- Fractal aggregates are very efficient to trap diffusing molecules

they are 'maze-like' porous materials

⇒ The apparent proportion of adsorbed gas molecules is **much larger** than expected from the theory

  - when $D_f \leq 2$ → because of the large specific surface
  - When $D_f > 2$ → because of the large specific surface + trapping



and there are consequences…

(*though not considered up to now…*)

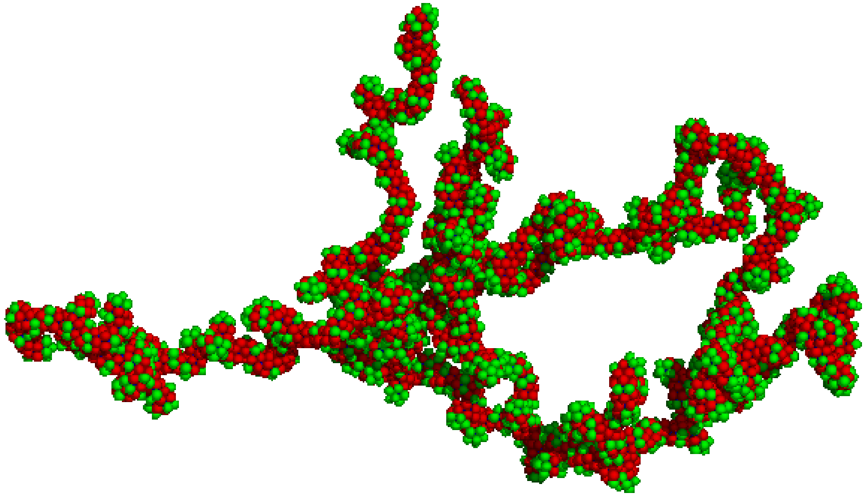
ANOMALOUS AD/ABSORPTION ON FRACTAL AGGREGATES AND CONSEQUENCES

- Fractal aggregates are very efficient to trap diffusing molecules

they are effective natural chemical reactors!

⇒ 1) when P/T large enough, aggregates appear as aggregates of **coated grains**

⇒ 2) **infrared radiation from vibrational modes** of the adsorbed gas molecules is more intense



do you want to detect special molecules? → *look where fractal aggregates are!*

SUMMARIZING…

- The information about the scatterers is mixed over the Stokes parameters functions

- It would be very much interesting to know analysis windows (*in the scattering angle, in the wavelength)* of the Stokes parameters, which carry simply relevant information. We know a few examples:
  - the Small-Angle-Scattering gives the value of fractal dimension and mean radius of gyration when scatterers are fractal aggregates with $D_f<2$
  - the absorption for light transmitted may give the size of aggregates

- … but many other examples are missing
  - for example: how can we deduce the value of the refractive index in a robust way?
  - adsorption of molecules lead easily to coated grains → which change in the Stokes parameters functions?
  - …