1. Aerosol Distribution Measurements by Laser - Doppler - Spectroscopy by J. BALDASSARI, France

ABSTRACT : Laser - Doppler - Spectroscopy is used to study particle size distribution, especially sodium aerosols, in the presence of uncondensable gases.

Theoretical basis are given, and an experimental technique is described.

First theoritical results show reasonnably good agreement with experimental data available ; this method seems to be a promising one.

I - INTRODUCTION

Light scattering techniques are applicable and frequently used in aerosol research. Almost all such methods are based on mean light intensity measurements and pose a number of inherent problems for sodium applications.

This paper describes a recent technique which will be extensively used in the aerosol research program undertaken by the French Atomic Energy Commission (CEA)

When a particle is illuminated by a laser beam, the scattered light frequency is shifted by a quantity Δf with respect to the incident light frequency.. If the particle is characterized by a Brownian motion, the frequency shift or Doppler frequency, depends on the speed of the particle and thus on its diffusion coefficient "D". By using the Stokes-Einstein relations the size distribution may be determined for the aerosol analyzed.

The method may be improved by using a real time correlator.

II - THEORETICAL CONSIDERATIONS

This section reviews the major theoritical aspects relevant to the experiments.

If we assume that $\vec{K}_o$ is the laser beam wave vector and θ the angle between $\vec{K}_o$ and the scattered wave vector $\vec{K}_s$ the field $\mathcal{E}_i$ scattered by the jth particle is, according to VAN DE HULST [1].

$$\mathcal{E}_j(t) = E_s \exp\left[i\left(\vec{K}\,\vec{r_j}(t) - \omega_o t\right)\right] \qquad (1)$$

Where :

- E_s : amplitude of the scattered field
- ω_o : incident light frequency
- $\vec{r_j}(t)$: position vector of the jth particle
- $\vec{K}$: scattering vector given by :

$$\vec{K} = \vec{K}_o - \vec{K} \qquad (2)$$

if $|\vec{K}_o| \simeq |\vec{K}_s|$ then $|\vec{K}| = 2|\vec{K}_o| \sin\theta/2$

since $|\vec{K}_o| = 2\pi/\lambda$ then $|\vec{K}| = \dfrac{4\pi}{\lambda} \sin\theta/2$ (3)

where : λ = wavelength of the laser beam.

The total scattered field detected by a receiver (the photomultiplier) is the sum of the $\mathcal{E}_i$ fields scattered by the N particles in the zone analyzed.

$$\mathcal{E}_s(t) = \sum_{j=1}^{N} E_s \exp\left[i\left(\vec{K}\,\vec{r_j}(t) - \omega_o t\right)\right] \qquad (4)$$

The autocorrelation function $\mathcal{R}_{\mathcal{E}}(\tau)$ of the scattered field takes the form :

$$\mathcal{R}_{\mathcal{E}}(\tau) = \overline{\mathcal{E}_s^*(t)\,\mathcal{E}_s(t+\tau)} \qquad (5)$$

where the bar indicates the mean time value and where * designates the conjugate complex.

For stationary ergodic phenomena [2] the functions $\mathcal{R}_{\mathcal{E}}(\tau)$ is independent of the time t. From the Wiener-Khinchine theorem [3] the signal frequency spectrum $\mathcal{S}(\omega)$ is related to the autocorrelation function $\mathcal{R}_{\mathcal{E}}(\tau)$ by

$$\mathcal{S}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} e^{i\omega\tau}\,\mathcal{R}_{\mathcal{E}}(\tau)\,d\tau \qquad (6)$$

After substituting relation (4) into equation (5), the autocorrelation function $\mathcal{R}_{\mathcal{E}}(\tau)$ may be written thus :

$$\mathcal{R}_{\mathcal{E}}(\tau) = E_s^2\, e^{-i\omega_o\tau}\, \overline{\exp\left[-i\vec{K}\,\overrightarrow{\Delta r(\tau)}\right]} \qquad (7)$$

$$\overrightarrow{\Delta r(\tau)} = \overrightarrow{r(t)} - \overrightarrow{r(t+\tau)}$$

Let $P(r,t\,/\,0,0)$ be the particle conditional probability distribution: $P(r,t/0,0)$ is the probability of finding a particle at point r at time t, if the same particle was at point r = 0 at time t = 0.

This probability $P(r,t\,/\,0,0)$ is equivalent to the probability $P(\Delta r,\tau)$ that a particle covers the distance Δr in the time Δt.

$P(\Delta r,\tau)$ is given by [4]:

$$P(\Delta r,\tau) = (4\pi D\tau)^{-1/2} \exp\left[-(\Delta r)^2/4D\tau\right] \qquad (8)$$

where D is the particle diffusion coefficient.

The mean value $\overline{\exp\left[-i\vec{K}\,\overrightarrow{\Delta r(\tau)}\right]}$ is written as :

$$\int_{-\infty}^{+\infty} P(\Delta r,\tau)\exp\left[-i\vec{K}\,\overrightarrow{\Delta r(\tau)}\right]d\Delta r \qquad (9)$$

By substituting relation (8) into (9) and integrating over Δ r, the relation $R_{\mathcal{E}}(\tau)$ becomes :

$$R_{\mathcal{E}}(\tau) = I_s\, e^{-i\omega_0\tau}\exp\left(-K^2D\tau\right) \qquad (10)$$

where $I_s = E_S^2$ and $K^2 = |\vec{K}|^2$

The frequency spectrum $\mathcal{S}_{\mathcal{E}}(\omega)$ for the scattered field, from (6) and (10) is :

$$\mathcal{S}_{\mathcal{E}}(\omega) = \frac{I_s}{\pi}\,\frac{K^2D}{(K^2D)^2+(\omega-\omega_0)^2} \qquad (11)$$

In order to obtain the frequency spectrum of the scattered light intensity I, the autocorrelation function $R_I(\tau)$ of the intensity I must be determined from the field autocorrelation function using relation [5]:

$$R_I(\tau) = I_s^2 + \left|R_{\mathcal{E}}(\tau)\right|^2 \qquad (12)$$

The frequency spectrum $\mathcal{S}_I(\omega)$ of the scattered light intensity is obtained by substituting relation (10) into relation (12) and taking the Fourier transform of (12) i.e.:

$$\mathcal{S}_I(\omega) = \frac{I_s^2}{\pi}\,\frac{2K^2D}{(2K^2D)^2+\omega^2} \qquad (13)$$

The spectra $\mathcal{S}_{\mathcal{E}}(\omega)$ and $\mathcal{S}_I(\omega)$ defined by relations (11) and (13) are centered on the laser frequency ω_0 and on the frequency $\omega = 0$ respectively .

It is therefore preferable to use the autocorrelation functions of intensity I (which yield spectra centered on the frequency $\omega = 0$) rather than the autocorrelation functions for the field $\mathcal{E}$ which give spectra centered on the laser frequency $\omega_0 \simeq 10^{14}$ Hz .

Another advantage of analizing the intensity frequency spectrum $\mathcal{S}_I(\omega)$ is that the half-width (HW) of the field spectrum is $HW = K^2D$ rad/s , while that of the intensity spectrum is $HW = 2K^2D$ rad/s

Thus the Laser-Doppler Spectroscopy method with light intensity correlation permits low frequency research while at the same time providing twice the resolution obtained with field measurements.

Allowing for (3), we obtain :

$$HW = \frac{16D\pi}{\lambda^2}\sin^2\theta/2 \quad Hz \qquad (14)$$

By using Stoke's relation :

$$D = kT/3\pi\eta d \qquad (15)$$

the diameter of the particles may be determined

By substituting (15) into (14) we obtain the system of curves shown in figure 1.

It may be noted that for particle diameters of less than $1\,\mu m$, the half-width becomes very significant. Thus, for example, when $\Theta = 90°$ and $d = 0.01\,\mu m$, the HW is approximately 4 500 Hz.

Consequently, small-diameter present a very strong Doppler signal This factor is potentially of great interest for the study of finely scattered aerosols.

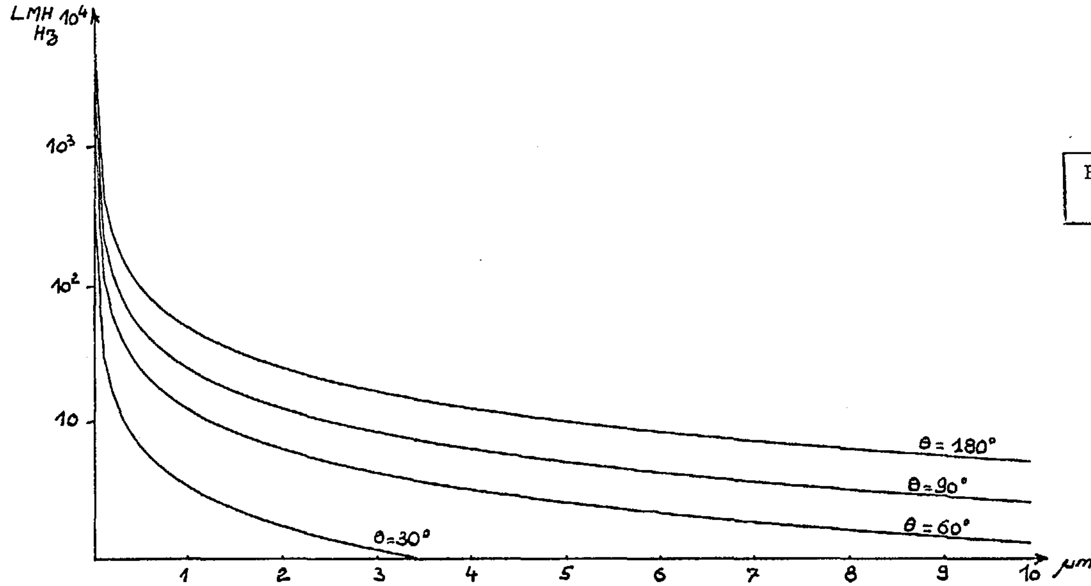

FIG. 1

III - EXPERIMENTAL CONDITIONS

Application of this technique to sodium aerosols is complicated by the nature of the medium (sodium aerosols in a neutral gas). the most important problem concerns the method's optical character: it is extremely difficult to keep the inspection windows clean in a sodium aerosol medium, and the measurment sequences must be performed very quickly to minimize this difficulty. For this reason the MALVERN measuring system [6] is perfectly suited to sodium aerosol research.

The major system components are show in figure 2.

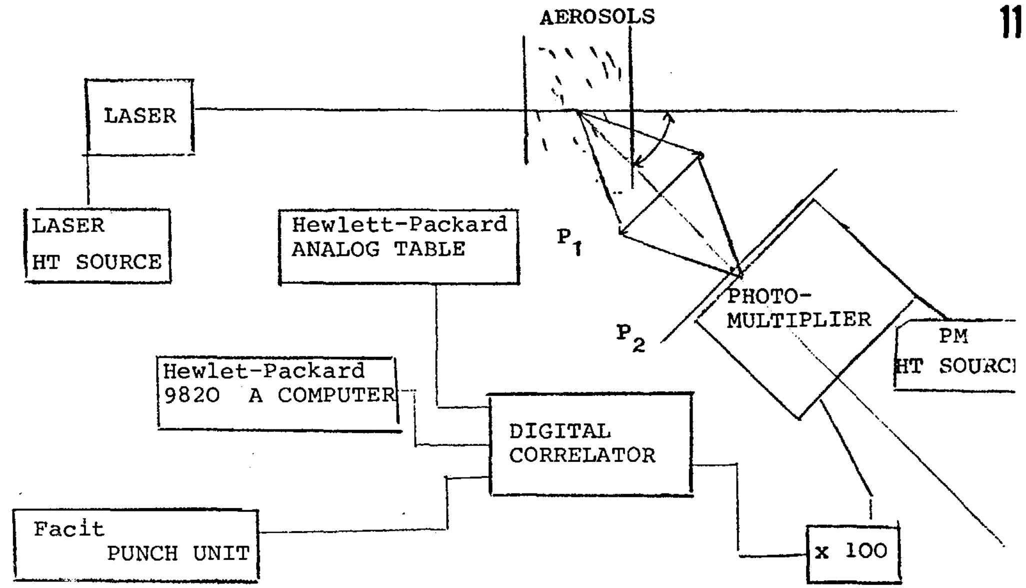

FIG. 2

The wavelength use is $\lambda = 4416$ Å at a power of 21 mW. The scattered light is picked up by a photomultiplier whose outup signal is processed by a x100 discriminator amplifier. The digital correlator the directly gives the scattered light intensity autocorrelation function $R_I(\tau)$. Correlator output may be directed to three units :

1) a Hewlett-Packard analog table
2) a Facit punch unit for processing on an IBM computer
3) a Hewlett-Packard 9820 A computer.

It is readily apparent that the correlator cannot compute the $R_I(\tau)$ autocorrelation function exactly : it simply calculates an estimator given by [7], [8] :

$$g_T^{(2)}(rT) = \frac{N \sum_N n_T(rT)\, n_T(0)}{\left[\sum_N n_T(0)\right]^2}$$

where :

- N : number of samples
- T : sampling time
- r : channel number
- $n_T(rT)$: number of photons counted during time T.

The complet correlation function is obtained by varying the rT "delay".

As it is often fastidious to compute the products $n_T(rT)N_T(0)$, the correlator may be programmed for "single clipped correlation" as described by a number of authors [9], [10]. The correlation function estimator is then given by :

$$g_k^{(t)}(rT) = \frac{N \sum_N n_k(rT)\, n(0)}{\sum_N n_k(0) \sum_N n(0)} \qquad (16)$$

where :

- k is the clip level
- n_k is such that (see figure 3)

$$n_k(t) = 1 \quad \text{if } n(t) \geq K$$
$$n_k(t) = 0 \quad \text{if } n(t) < K$$

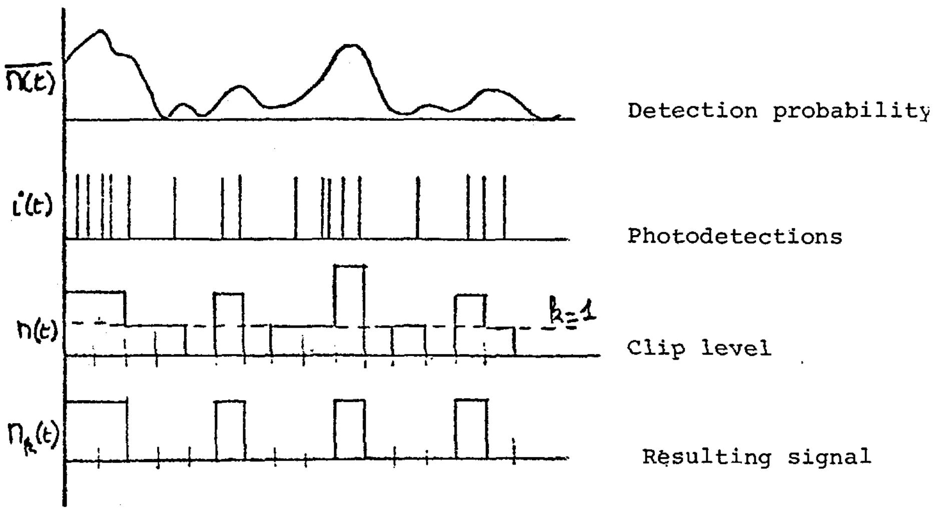

FIG. 3

IV - EXPERIMENTAL FINDINGS

An experimental research program is currently in progress at the CADARACHE Fast Neutron Reactor Department. The initial results have been encouraging despite the difficulties related to the sodium aerosol medium. Figure 4 shows a typical scattered intensity correlation function obtained.

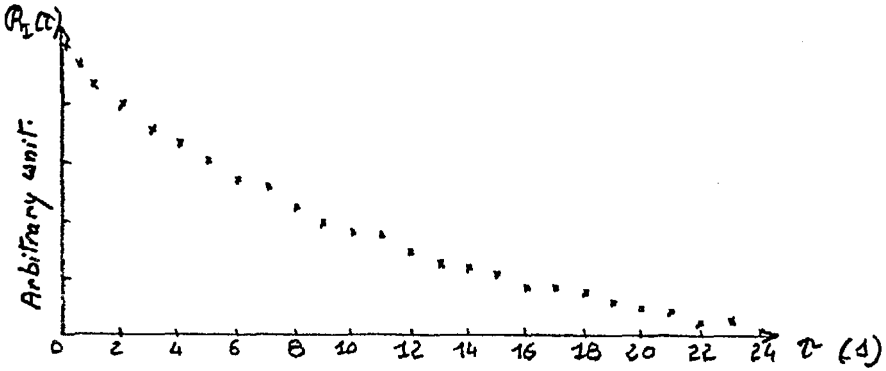

FIG 4.

Allowing for the experimental conditions, the mean diameter for this sample may be estimated as 4 μm.

V - CONCLUSION

The LDS method for determining aerosol size distributions is perfectly suited to sodium aerosols. The satisfactory accuracy of the method permits very rapid measurements to be made with the MALVERN measuring system.

Other LDS applications [11], [12] may be adapted for sodium aerosol research.

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NOMENCLATURE

- D Particle diffusion coefficient
- d Particle diameter
- E_s Amplitude of scattered field
- $g_R^{(2)}(\cdot)$ Clipped intensity correlation function
- $g_T^{(2)}(\cdot)$ Unclipped intensity correlation function
- I_s Average intensity
- i Such that $i^2 = -1$
- $\vec{K}$ Scattering vector
- $\vec{K_0}$ Laser wave vector
- k Boltzmann constant
- N Number of sample
- $n_T(\cdot)$ Number of counted photons
- $P(\cdot,\cdot/\cdot,\cdot)$ Probalility distribution
- r POsition vector
- $\vec{r_j}(\cdot)$ Position vector of the jth particle
- rT Delay time
- T Absolute temperature in °K
- t Time
- $\bar{\alpha}$ Average of α
- α^* Complex conjugate of α
- η Viscosity
- θ Angle of scattering
- λ Wavelength
- ω Frequency
- ω_0 Laser frequency
- τ correlation time
- $\mathcal{E}_j$ Field scattered by the jth particle
- $\mathcal{E}_s$ Total scattered field
- $R_I(\cdot)$ Field autocorrelation function
- $\mathcal{F}(f)$ Fourier spectrum of function f
- $\mathcal{F}_{\mathcal{E}}(\cdot)$ Fourier spectrum of scattered field