Self-aligning optical particle sizer for the monitoring of particle growth processes in industrial plants

A. Bassini, S. Musazzi, E. Paganini, and U. Perini
CISE Tecnologie Innovative, via Reggio Emilia 39, 20090 Segrate, Italy

F. Ferri
Istituto di Scienze Matematiche, Fisiche e Chimiche and INFM (Istituto Nazionale di Fisica della Materia), University of Milan at Como, via Lucini 3, 22100 Como, Italy

(Received 4 December 1997; accepted for publication 23 March 1998)

We describe a diffraction based optical particle sizer to be used on-line in an industrial plant for monitoring the growth process of polystyrene beads in the range (400 μm–4 mm). The instrument has been designed to perform elastic light scattering measurements at very small angles (from 8 × 10^{-5} to 8 × 10^{-3} rad) and is provided with an active servo system that controls the beam alignment during operations. © 1998 American Institute of Physics.

I. INTRODUCTION

Optical particle sizers are one of the most important tools so far utilized for the nonintrusive characterization of polydisperse samples of particles. Under the hypothesis of having a sample made by nominally spherical particles, optical particle sizers provide a "quasi real time" measurement of the particle size distribution. Their principle of operation is well known and relies on the analysis of the angular distribution of the light scattered by the sample at low angle in the forward direction. Solid and liquid particles (sprays) can be analyzed in a wide range of sizes (from the micron to the millimeter region) provided that the indexes of refraction of both the sample and the surrounding medium are different.

Commercially available instruments of this type exist (since the early 1980s) and they are currently used in a large number of laboratories both for scientific and industrial applications. However, typical features of commercial equipments, such as their poor modularity as well as their vulnerability to external stresses (both thermal and mechanical), make them not very suitable for on-line applications in industrial plants. In many cases, therefore, to meet the requirements stemming from the specific application, custom-made instrumentation capable of operating in hostile environments should be developed.

An example is reported in this article where we present a prototype of an optical particle sizer to be used for the control of the growth process of spherical polymer aggregates (polystyrene beads) in an industrial plant. Particles, whose mean diameter grows from few tens of microns to some millimeters during the whole process, are drawn from the reactor and brought to the test cell via a suitable sampling line. Monitoring of their mean diameter has to be done at time intervals of about 3 min at the beginning of the growth process and at larger time intervals during the following phases.

Due to the peculiar measuring range, the instrument design is very critical. The scattered light, in fact, has to be detected at extremely small angles, over an angular range of two decades (from 8 × 10^{-5} to 8 × 10^{-3} rad). As it can easily be understood, to guarantee the requested alignment accuracy during the whole measurement time, design requirements in terms of mechanical and optical stability are very high. We coped with this problem by providing the instrument with an active servo system that controls the alignment during operations.

Experimental tests will be presented which demonstrate the compensating capability of the alignment control loop as well as the ability of the system to correctly recover particle size distributions in the foreseen measuring range.

II. PRINCIPLE OF OPERATION

The basic concept of optical particle sizes hinges on the measurement of the angular distribution of the light elastically scattered by the sample. Once the scattered intensity distribution is known, the sample size distribution can be determined via a suitable inversion algorithm. The optical scheme is quite simple. A parallel beam of light is sent to the test cell, the scattered light is collected by a Fourier lens and forwarded to a sensor placed in the lens focal plane. The sensor is an array of annular photodiodes with a tiny hole in the center. Thus each sensor element collects the light scattered at a given angle, while the transmitted beam, being focused into the tiny hole, passes clear the sensor without affecting the measurement. The above scheme is a typical quasi-exact Fourier transform arrangement, since the electric field distribution on the sensor plane, apart from an additional phase factor, is the Fourier transform of the test cell transparency function. The associated intensity distribution (which is the quantity actually measured), however, is correct since it does not depend on the field phase distribution.

For spherical particles much larger than the illuminating wavelength (as it is our case), the low-angle elastically scattered intensity distribution is independent of the particles refractive index and can be approximated by the well-known Airy function \( I(q,a) = C \alpha^2 \left[ \frac{J_1(qa)}{qa} \right]^2 \),

\( \text{(1)} \)
where \( a \) is the particle radius, \( C \) is an arbitrary constant independent of \( a \), \( J_1 \) is the Bessel function of order one, and \( q \) is the wave vector given by

\[
q = \frac{2\pi r}{\lambda f} \tag{2}
\]

being \( r \) the radius coordinate in the sensor plane, \( f \) the Fourier lens focal length, and \( \lambda \) the illuminating wavelength. Equation (2) has been derived by approximating \( \sin \theta \sim \theta \) (being \( \theta \sim r/f \) the scattering angle) and by taking into account refraction effects due to the scattering cell walls. Note that the increase of the scattering angle due to refraction effects is cancelled out by the reduction of the laser wavelength in the medium, leading to an expression of \( q \) independent of the medium refractive index.

Since in real situations more particles are present in the test cell and their relative positions vary randomly during the measuring time, the resulting time-averaged intensity distribution in the sensor plane will be the sum of different contributions, each of them corresponds to the Airy diffraction pattern generated by single particles in the sample. Thus, the total scattered intensity \( I_{TOT}(q) \) is given by the first-kind Fredholm integral equation

\[
I_{TOT}(q) = \int I(q,a)N(a)da, \tag{3}
\]

where \( N(a) \) is the particle number distribution and \( I(q,a) \) is the kernel provided by Eq. (1). Equation (3) is a typical example of an ill posed problem, that is, in the presence of noisy data rather different distributions can fit the experimental data to the same level of accuracy. Consequently, the solutions might be highly unstable and unreliable, and it becomes of crucial importance to adopt a suitable inversion algorithm. The choice of the algorithm determines the measuring range, i.e., the range of radii over which the particle size distribution should accurately be retrieved. Typically, in the case the scattered light is detected within a wave vector range \( (q_{\text{min}}, q_{\text{max}}) \), the inversion of Eq. (3) is carried out over a range of radii \( (a_{\text{min}}, a_{\text{max}}) \sim (1/q_{\text{max}}, 1/q_{\text{min}}) \). It should be pointed out, however, that this is only a rough indication of such a range since its extension actually depends on many factors, such as the detection angular accuracy, the noise level present on the recorded data, and, as mentioned above, the adopted inversion algorithm. All these boundary conditions may account for correction factors as large as 2 (or even more) in the estimate of \( a_{\text{min}} \) and \( a_{\text{max}} \). In our case, the inversion of Eq. (3) was carried out by using a recently proposed nonlinear inversion algorithm. Computer simulations reported in that work show that the inversion algorithm is able to recover the particle size distribution over a range given by

\[
(a_{\text{min}}, a_{\text{max}}) \sim (q_{\text{max}}^{1.5}, q_{\text{min}}^{1.5}) = \left( \frac{0.24\lambda f}{r_{\text{max}}}, \frac{0.24\lambda f}{r_{\text{min}}} \right), \tag{4}
\]

where \( r_{\text{min}} \) and \( r_{\text{max}} \) are the innermost and outermost average radii of the annular sensor elements. Taking into account that for \( qa\sim1.5 \), the Airy function decays at a value which is approximately a factor of 2 lower than its maximum, the left-hand side of Eq. (4) provides two different measuring conditions: (i) the smallest particle radius \( a_{\text{min}} \) should be large enough to generate a diffraction pattern whose intensity distribution decays (at least) by a factor of 2 over the entire wave vector range; at the same time, (ii) the radius \( a_{\text{max}} \) of the largest particle in the sample should be small enough so as to generate a diffraction intensity distribution that decays at \( q_{\text{min}} \) by (no more than) a factor of 2 with respect to its maximum. The right-hand side of Eq. (4) shows also that the measuring range depends on the system geometry, i.e., on \( r_{\text{min}}, r_{\text{max}}, f \) as well. Whereas the radii \( r_{\text{min}} \) and \( r_{\text{max}} \) account for the dynamical extension of the range of recoverable radii \( (a_{\text{max}}/a_{\text{min}}, r_{\text{max}}/r_{\text{min}}) \), it is the focal length \( f \) which determines the absolute scale of such a range. For a given array sensor geometry, the larger is the average size of the particles to be analyzed, the longer is the required Fourier lens focal length. In our case, due to the unusually large particle size (millimeters), the scattered light must be detected at very low wave vectors and, therefore, a remarkably long focal length has to be used. As it will be discussed in Sec. III, this implies that \( f > 2 \text{ m} \).

III. EXPERIMENTAL SETUP

A. Optical layout

The general layout of the experimental setup is schematically shown in Fig. 1. The instrument is made by two separate units: the projection unit and the receiving unit. The projection unit is used to generate the illuminating probe beam. The light source is provided by a linearly polarized 5 mW HeNe laser, with \( \lambda = 0.6328 \ \mu \text{m} \) and a beam waist at \( 1/e^2 \) of 0.8 mm. The beam is spatially filtered and made divergent by means of a spatial filter (SF) assembly made by a \( 5 \times 5 \) microscope objective (\( f = 25 \text{ mm} \)) and a 45 \( \mu \text{m} \) diameter pinhole that can be accurately positioned in the microscope objective focal plane. After reflection onto the aluminum coated plane mirrors M1 and M2 (\( \lambda/10 \) flatness), the beam is collimated by the lens L1 (achromat doublet, 150 mm diameter, 2250 mm focal length) and sent to the exit optical window OW1 (150 mm diameter, \( \lambda/4 \) flatness). The need of having an optical window at the exit/entrance of the two units is determined by the fact that, for safety reasons, each unit has been mounted in a sealed box filled with inert gas. The window OW1 is slightly tilted with respect to the

![Fig. 1. Schematic diagram of the experiment setup.](image-url)
optical axis so to allow the reflected portion of the beam to be focused (by the lens L1) back to a photodiode (not shown in the figure) which is used to monitor the laser power during tests. The parallel beam emerging from the projection unit (that has a diameter at 1/e² of 75 mm) is the probe beam used to shine the sample flowing through the test cell (TC) to be described later on. Both the transmitted beam and the scattered light contributions are collected by the receiving unit through the entrance window OW2 and the lens L2 (which are identical to OW1 and L1, respectively). The two contributions are brought to the array sensor placed in the focal plane of L2 via reflections onto different optical elements, i.e., the two mirrors M3 and M4 (identical to the mirrors M1 and M2, respectively) and the wedged (8° wedge) aluminum-coated beam splitter plate BS (95% reflectivity). To reduce stray light contributions, the wedge has been properly determined so to minimize, by total internal reflection, undesired reflections from the second beam splitter surface. After the three reflections, the scattered light is detected by the array sensor AS, while the focused transmitted beam, passing clear through the pinhole drilled in the center of the array sensor, is detected by the photodiode PH and is used to monitor the sample turbidity. The attenuated replica of the transmitted beam, emerging from the beam splitter BS, impinges onto the position sensing device PS (a quadrant photodiode detector, UDT, mod PIN-Spot/9D) whose center is optically conjugate with the annular array sensor central pinhole. The signals out from the position sensing device are fed to the servo unit which controls the angular position of the tilting mirror M4 via two piezoelectric actuators. In this way, by locking the replica of the transmitted beam onto the center of the position sensing device, the servo guarantees that proper alignment is also maintained in the measuring arm. Thus, the transmitted beam is correctly focused into the array sensor center pinhole and does not affect the measurement with undesired light contributions on the innermost array sensor elements.

The array sensor AS is a custom-made detector manufactured by Centronic on our design. It consists of 31 separate concentric annular quarters of rings alternatively located on the opposite sides of a vertical axis passing through the sensor center (see Fig. 2). Except for tiny gaps between the sensitive elements, the whole area of the sensor is sensitive, providing a detecting filling ratio of almost 100%. The mean radii and thicknesses of the rings have been chosen according to an almost geometrical progression of ratio equal to about 1.16, therefore providing the same relative angular accuracy in the detected scattered light. In particular, with the exception of the innermost array elements, due to the peculiar geometry of the sensor, this accuracy is equal to δr/ r = δθ/θ = 0.25. The pinhole in the center of the sensor has a diameter of 200 μm, while the innermost and outermost array sensor elements have, respectively: mean radius \( r_{\text{min}} = 0.185 \text{ mm} \) and \( r_{\text{max}} = 17.322 \text{ mm} \), and thickness \( t_{\text{min}} = 80 \text{ μm} \) and \( t_{\text{max}} = 4.655 \text{ mm} \). It should be noticed that: (i) the detectable angular dynamic range \( (\theta_{\text{max}})/\theta_{\text{min}}) \), which is equal to the ratio \( r_{\text{max}}/r_{\text{min}} \), is of about two orders of magnitude; and (ii) the ratio between the outermost sensor element area (1.27×10⁸ μm²) and the innermost sensor element area (3.23×10⁴ μm²) is about 5×10³. This allows to get a higher sensitivity in the detection of light scattered at larger angles (where the intensity might be very low) and, at the same time, to reduce the overall dynamic range of the signals out of the array sensor elements.

As anticipated above, for a given sensor geometry, the Fourier lens focal length actually determines the measuring range. Thus, by using Eq. (4) and recalling that the particle diameters of interest are in the range (40 μm–4 mm), it comes out that the Fourier lens focal length has to be slightly larger than 2 m. Among commercially available achromat doublets with such a long focal length and reasonably large diameter, we selected the \( f = 2250 \text{ mm} \), \( D = 150 \text{ mm} \) precision achromat doublet manufactured by Spindler and Hoyer. By using this lens the scattered light can be analyzed over the angular range 8×10⁻⁵–8×10⁻³ rad.

Let us now briefly describe how the diameter \( d_G \) of the collimated beam impinging onto the test cell was actually determined. The leading criterion was to minimize the focused transmitted beam intensity distribution in correspondence of the innermost array sensor elements. As is known, in fact, when a Gaussian beam is focused by an aberration free lens with a finite pupil diameter, the focal plane intensity distribution depends on the actual illuminating f-number \( (f^d = f/\alpha \text{ where } f \text{ is the lens focal length and } d_G \text{ is the width at } 1/e^2 \text{ of the parallel Gaussian illuminating beam) as well as on the diffraction effects due to the finite lens diameter } D \text{. In the case of an ideal lens with } D \gg d_G \text{, such a distribution has a Gaussian profile with a beam waist given by } 2\omega_0 = 1.27\lambda f^d \text{. However, when the lens has a finite aperture } D \text{ comparable with } d_G \text{, truncation effects take over and circular diffraction rings appear around the central bright spot therefore generating undesired background signals on the innermost array sensor elements. The illuminating beam diameter } d_G \text{ should therefore be large enough to generate a diffraction limited beam spot } 2\omega_0 \text{ much smaller than the array sensor central pinhole, and at the same time, it should be small enough to avoid truncation effects. To get the right compromise between these two opposite requirements, we computed the intensity of the light diffracted on the innermost array sensor element as a function of the truncation ratio } \alpha = D/d_G \text{. Our results show that, in the range } \alpha \gg 3 \text{, the detected intensity monotonically decreases and it is always comparable with that one would expect to measure in}
the case of an unperturbed Gaussian beam. It attains a minimum for $3 > \alpha > 2$ and then starts to increase very rapidly, reaching, for $\alpha = 1.5$ a level which is approximately three orders of magnitude larger than the one measured at $\alpha = 3$. According to these results, a reasonable compromise is $\alpha = 2$, which corresponds to the situation of having a collimated beam diameter $d_G = 75$ mm and an $f$/number $f^d = 30$. The diffraction limited beam spot is, in this case, much smaller ($2w_0 = 24 \mu m$) than the array sensor pinhole diameter ($200 \mu m$), therefore indicating that the choice of the Fourier lens was fairly appropriate.

As a final comment, we would like to show a further consequence in having a small beam spot diameter. Let us consider the optical noise associated to the average intensity $\langle I \rangle$ detected by each element of the array sensor. It is due to the finite number of speckles falling onto the sensitive area of each element. By denoting with $\langle N_i \rangle$ the average number of speckles collected by each element, the (fractional) optical noise $\delta I/\langle I \rangle$, for $\langle N_i \rangle \gg 1$, is given by $\delta I/\langle I \rangle \sim (\langle N_i \rangle)^{-0.5}$. The speckle grain noise is therefore fractionally smaller for the outermost sensor elements than for the innermost ones. By taking into account that the average speckle size on the array sensor plane is, in the absence of aberrations, equal to the diffraction limited beam spot diameter ($\sim 24 \mu m$), the average number of speckles on the outermost and innermost elements are $\sim 2.5 \times 10^5$ and $\sim 50$, respectively. The corresponding noise levels are $\delta I/\langle I \rangle \sim 0.2\%$ and $\sim 14\%$, respectively. In our case, the speckle grain noise has been reduced by averaging the measured intensity distribution over many different configurations of the speckle field (see Sec. III C).

B. Mechanical layout

A photo of the experimental apparatus is shown in Fig. 3. As it can be noticed both the projection and the receiving unit optical components are mounted on two identical standard honeycomb aluminum plates ($1200 \times 600 \times 80$ mm$^3$) with a nominal end-to-end flatness of $\pm 0.12$ mm. The two plates are clamped on the bottom of two sealed metallic boxes (not shown in the figure) via distortion free cinematic mounts (the standard “cone, grove, and flat” mount) that guarantee the requested mechanical stability. The two sealed boxes, that are supported by four nonisolating legs, are connected by a pipe and can be filled with an inert gas (argon) to get the instrument compliant with the antideflagration safety rules.

Apart from the tilting mirror support, which is a standard Burleigh gimbals mount properly modified to allow the use of PZT actuators, all the other optical mounts are custom made. To minimize stresses caused by mechanical fasteners (that would eventually corrupt optical performances), mirrors as well as lenses are held in position in their mounts by means of adhesive silicon elastomers of adequate flexibility. Care has also been payed in the design of optical subsystems/assemblies to prevent misalignments that cannot be compensated by the active servo system and would eventually cause a malfunctioning of the instrument.

One of the most critical items is the spatial filter assembly. To get the requested stability, a compact springless (nominal slack-free) spatial filter mount has been manufactured where the microscope objective and the pinhole mechanical supports can be locked after the alignment procedure. In this way, the spatial filter alignment is maintained also in the presence of severe thermal and mechanical stresses. To guarantee proper alignments between the incoming laser beam and the microscope objective, the spatial filter assembly is directly clamped on the laser support.

A second critical item is the beam splitting block. In order not to loose the optical conjugation between the position sensing device and the multielement array sensor centers, the wedged beam splitter plate as well as the two conjugate elements should not change their mutual positions during operations. To get the requested mechanical stability, the three components are rigidly mounted on a unique base-plate made by low-expansion material (invar) which is properly clamped on the honeycomb plate.

The test cell (see Fig. 4) where the sample is flowing, is supported by a rack and is positioned between the projection and the receiving units. It is a stainless-steel vessel provided with two optical windows ($150$ mm diameter, $\lambda/4$ flatness). It is made by three separate parts: the main body and two interface sections for inflow and outflow tubing connections.
The main body consists of a hollow structure where two optical windows are mounted (by means of fixation flanges) so to provide optical access to the streaming particles. Test particles flow through the main body in a rectangular (6 \times 146 \text{ mm}^2) channel. To have a uniform particles stream (so to fill homogeneously the test region), the particle laden fluid should match as close as possible a laminar flow regime. This condition has been met by proper tapering the inlet and outlet channels in the interface sections. The two channels, that have a smoothly varying rectangular cross section of constant area and variable aspect ratio, fit a (30 \times 30 \text{ mm}^2) square aperture on one side and a (6 \times 146 \text{ mm}^2) rectangular aperture on the opposite side. The two interface sections are also provided with an additional fitting block for the connection between the (30 \times 30 \text{ mm}^2) square aperture and the incoming/outcoming circular pipes.

The velocity of the fluid in the cell is about 0.5 m/s. This means that the typical residence time (i.e., the time requested to a particle to cross the test region, or in other words, the time needed for replacing the set of scattering particles with a new one) is of the order of 0.3 s. Since this time interval corresponds to the typical speckle fluctuation time, it represents an important parameter for the definition of the measuring time.

C. Electronic layout

A block diagram of the electronic units that provide both data acquisition and alignment control capabilities is shown in Fig. 1.

Let us first describe the design criteria relevant to the data acquisition electronic chain. They can be derived from the main features (amplitude, dynamic range, and time constant) of the optical signals to be detected. Since optical signals are generated by the scattering of light from particles much larger than the wavelength, whose concentration can be rather arbitrarily set, their amplitudes are usually quite high and there is no need of a high sensitivity detection system. Somewhat more demanding design criteria stem from the dynamic range requirements. Indeed, due to the quite large particle diameters range, the signal sequence out from the array sensor elements may span over several orders of magnitude. A rough estimate of the requested dynamic range can be obtained by considering the worst operating condition, i.e., when samples of very small particles are to be investigated. In this case, the array sensor is illuminated by a rather uniform intensity distribution and the detected signals scale approximately as the elements active areas (the ratio between the outermost and innermost array sensor elements active area being \( \sim 5 \times 10^3 \)). By considering that the typical sample attenuation during measurements is of the order of 5\%–10\%, to get an accuracy of a few percent, the overall dynamic range should be \( \sim 10^6 \). Finally, we remember that the time scale at which changes of the optical signals occur due to speckle fluctuations is, as described above, fairly slow (\( \sim 0.3 \text{ s} \)).

On the basis of the above-mentioned requirements, the electronics design as well as the selection of the electronic components was quite straightforward and only a brief description is reported here. The current signals out from the 31 array sensor elements and from the photodiode PH are converted into voltage signals by means of the preamplification transimpedence stage (TS) positioned close to the detectors. The 32 analog signals coming from the transimpedence stage are fed to a 12-bit computer controlled A/D conversion unit located in the main electronic box. Since the sampling rate of the A/D unit is 10 kHz, each channel is sampled at a rate of about 300 Hz. This oversampling with respect to the intrinsic bandwidth (3 Hz) of the optical signal makes it possible to average over the electronic noise. To average over the actual optical signal fluctuations the overall measuring time was about 30 s (which is in any case reasonaaly smaller than the typical sampling time requested to control the polystyrene growth process). In this way each channel is sampled \( 10^4 \) times. This corresponds to average over \( \sim 100 \) statistically independent samples so to reduce by a factor of 10 the standard deviation of the optical signals fluctuations. By taking into account that the rms amplitude of the optical noise (which depends on the number of coherence areas collected by each sensor element, see Sec. III A) is, in the worst case, of the order of 14\%, a reduction by a factor 10 of the standard deviation is enough to obtain measurements accurate within a few percent.

Because of the requested dynamic range of \( 10^6 \), the A/D
unit is provided with a software programmable gain amplifier that allows to set (according to a geometrical progression of ratio 2) the gain of each channel in the range 1–128. Digital signals are brought to the remote computer, which is about 40 m distant from the measuring units, via a low noise 4–20 mA current driver (the transmission unit T1 in Fig. 1). The computer is a standard PC equipped with a digital I/O card. Dedicated software has been properly developed both for the instrument control and for data handling.

Let us now describe the electronics of the optical alignment control loop. It has been designed according to the following requirements: (i) the alignment controller has to be free of any offset errors so to guarantee that, under steady state conditions, the alignment could be reached and maintained over long periods of time; and (ii) since misalignments are mainly caused by temperature-induced mechanical instabilities, the requirements on the control loop are fairly mild and the response time can be rather slow (seconds). Thus we exploited a purely integral servo loop. This type of controller in fact guarantees a real zeroing of the misalignment errors signals and provides a very long term stability since its response time can be set much longer than the uncontrolled system response time. In our case, the latter time is determined mainly by mechanical inertia and is of the order of a few tenths of a second.

As described in Sec. II, the alignment control loop is based on the optical conjugation between the array sensor central pinhole and the center of the position sensing device (see Fig. 1). Thus, when the transmitted beam is perfectly centered on the position sensing device, it is also correctly focused onto the array sensor pinhole. Misalignments are revealed by detecting the four current signals, $s_1$–$s_4$, out of the quadrant detector. The four signals are fed to a custom-made circuit which generates the two voltage error signals along the horizontal and vertical axis, $\Delta x$ and $\Delta y$. They are given by

$$\Delta x = \frac{(s_1 + s_3) - (s_2 + s_4)}{s_1 + s_2 + s_3 + s_4},$$

$$\Delta y = \frac{(s_1 + s_2) - (s_3 + s_4)}{s_1 + s_2 + s_3 + s_4},$$

where the subscripts 1, 2, 3, and 4 refer to the quadrant position (being 1 the top-left quadrant and assuming that the others are arranged in a clockwise sequence). The electronics was tailored so to have a maximum swing of the error signals of $\pm 0.5$ V. The error signals are fed to two low drift offset integrator circuits characterized by an integration time of 1 s. Each output of the integrators is summed to a constant reference voltage of 0.5 V and fed to the input of a low noise voltage amplifier (with a fixed gain equal to 100). The outputs of the amplifier are therefore ranging between 0 and 100 V (50 V being the initial setpoint). These are used to drive the piezoelectric actuators that control the angular orientation of the tilting mirror (M4). The piezoelectric actuators (PI, mod. P-840.60) have an expansion coefficient of $0.9 \mu m/V$ and a maximum elongation of $90 \mu m$. By taking into account that the actuators are positioned at a distance of 50 mm from the center of rotation of the gimbals mount, rotations of the tilting mirror are controlled with an angular sensitivity of $\sim 1.8 \times 10^{-5}$ rad/V. Since the incoming beam is deflected by an angle which is twice the mirror rotation angle, the corresponding linear sensitivity in the array sensor plane (which is 500 mm distant from the tilting mirror) is $\sim 18 \mu m/V$. The overall compensating capability is therefore $\pm 0.9$ mrad on the tilting mirror and $\pm 0.9$ mm on the array sensor plane. This corresponds to compensate for a maximum angular deviation of the collimated laser beam impinging on the test cell of approximately $\pm 0.2$ mrad.

IV. MEASUREMENT PROCEDURE AND DATA HANDLING

A measurement of the angular distribution of the light scattered by the sample is performed in two steps. First, we record the so-called background signals (i.e., the signals mainly due to stray light contributions) by filling the test cell with water only. These signals are normalized by the laser incident power and corrected for the dark current offsets of each array sensor element. Second, we record the sample signals by filling the test cell with the sample and by repeating the same procedure. In the two measurements the cell is kept in the same position, and this is fundamental in order to correctly subtract the background signals. Then, since both the scattered and the stray light are attenuated by the same factor when passing through the scattering sample, the background signals are reduced by a factor equal to the beam attenuation, and subtracted from the sample signals. The scattered intensity distribution is then obtained by normalizing the background-subtracted signals for the gains corresponding to each channel. The values of these gains are given by the product of the sensor element active area times the transimpedance of the corresponding current–voltage converter. Since the nominal value of the sensor elements active areas may be somewhat inaccurate (it depends on the particular array sensor being used), at least at small angles where these areas are very small, a calibration procedure is recommended. This can be carried out rather easily by using pinholes, as described in Sec. V B.

Once the scattered intensity distribution is obtained, the particle size distribution is recovered by inverting the intensity data by means of the nonlinear inversion algorithm described in Ref. 8. As described in this reference, the recovered particle size distribution is approximated by a histogram with a number of classes equal to the number of experimental data (31 in our case). The range of diameters over which the inversion was carried out was determined according to Eq. (4) and was 40 $\mu m$–4 mm. Within this range, the classes of the histogram were chosen so that their average diameters and thicknesses scale according to a geometrical progression, so that all of them have the same relative width of approximately 16%.

V. PRELIMINARY TESTS

This section describes the main tests carried out on the instruments and discusses some of its performances and limitations. The instrument was tested under conditions matching as close as possible those for which it has been designed.
A. Stability tests

The beam pointing stability has been tested both in the absence and in the presence of the alignment control loop. Tests have been performed by varying the room temperature and by recording, as a function of time, the incident and transmitted laser power, the signals out of the two innermost array sensor elements and the room temperature. The test was repeated several times under different room temperature conditions. Figures 5 and 6 give an example of these tests.

Figure 5 shows the changes in the room temperature, the transmitted beam power variation, and the signals variations out from the two innermost array sensor elements as function of time over a period of ~17 h.

FIG. 5. Stability test of the instrument optical alignment. The measurements were carried out with the alignment servo loop switched off. (a) The room temperature, (b) the transmitted beam power variations, and (c) the signals variations out from the two innermost array sensor elements are reported as function of time over a period of ~17 h.

B. Test with pinholes

The proper functioning of the instrument and its ability to correctly recover the scattered intensity distribution over extreme small angles, was tested by using circular pinholes of different diameter (from 400 μm to 2 mm). For each diameter, we prepared a mask made of a 2 mm thick brass plate drilled with a precision drill press. The holes (of the same size) were randomly distributed over a circular area of about 100 mm in diameter and their number has been maxi-
mized so as to get the highest signals before reaching the saturation level. The pinholes diffraction pattern was measured by placing the mask on the optical axis in correspondence to the test cell plane. Data have been recorded without background subtraction. Figure 7 shows the diffraction pattern generated by the 400 μm pinholes, plotted as a function of the wave vector $q$. Data have been rescaled by an arbitrary constant so to overlap with the theoretical curve at low $q$ vectors. The latter one was determined by averaging the Airy function over the array sensors areas. The figure shows that experimental data (that span over three orders of magnitude) match the theory fairly well over the entire range of wave vectors. When larger pinholes are used, the intensity dynamic range becomes even larger. As shown in Fig. 8, where we report $I(q)$ vs $q$ for 2 mm pinholes, the intensity varies over more than five decades. Also in this case, matching between theory and data is quite remarkable over the entire $q$ range. For both Fig. 7 and Fig. 8, it should be noticed that it is possible to distinguish only few secondary maxima in the diffraction pattern. This is due to the fact that the sensor elements have a finite non-negligible thickness which limits the resolving capability and causes a smearing of the Airy function secondary minima.

As a final comment, it should be pointed out that the use of pinholes for testing the instrument has several practical advantages. First, it can be used to correct the relative gains between different channels of the sensor, therefore providing an easy way to calibrate the entire instrument. Second, being the total power diffracted by the pinholes per unit solid angle a known quantity, the instrument can be calibrated on an absolute scale of intensities. Although an absolute calibration may sometimes be quite important (since many information can be inferred from absolute intensity measurements), it has not been carried out being this task beyond the purpose of this work. Third, the use of circular apertures of known size provides a precise and accurate tool for testing the inversion algorithm utilized for recovering the sample particle size distribution from the measured intensity distribution.

FIG. 7. Diffraction pattern generated by a set of 400 μm diameter pinholes. Comparison between experimental data (open circles) and the theoretical curve obtained by averaging the Airy function over the sensor elements areas.

FIG. 8. Diffraction pattern generated by a set of 200 μm diameter pinholes. Comparison between experimental data (open circles) and the theoretical curve obtained by averaging the Airy function over the sensor elements areas.

C. Test with latex particles

The instrument has been tested on real samples by studying aqueous suspensions of calibrated polystyrene spheres. We used particle samples (from Duke Scientific Corporation) with nominal diameters ranging between 5 and 40 μm. The sample monodispersity, that has been determined by means of an optical microscope, is fairly good with standard deviations as small as a few percent. All the samples have been diluted in distilled water (previously filtered through a 0.22 μm Millipore membrane) so as to get a ~5% attenuation of the probe beam on a 0.5 cm thick test cell (a standard cell for photometry).

Since test particles are much smaller than the polystyrene beads that are of interest for this work, the measuring range has proportionally been reduced by positioning the test cell much closer to the array sensor (between the mirror M4 and the beam splitter BS in Fig. 1) in the region where the probe beam is convergent. Since, in the case the cell thickness is much smaller than the cell to sensor distance, the parallel and the converging beam illuminations schemes are equivalent, the measuring range can still be determined by means of Eq. (3) provided that the lens focal length $f$ is replaced by the cell to sensor distance $d$. In our case we used $d=150$ mm, corresponding to a wave-vector range of $1.2 \times 10^2$–$1.2 \times 10^4$ cm$^{-1}$ and to a diameter measuring range of 5–50 μm.

Measurements have been performed in two steps, by following the procedure described in Sec. IV. The scattered intensity distribution $I(q)$ is reported in Fig. 9 as a function of $q$ on a log-log plot. As one can notice, data exhibit a rather flat intensity distribution at low angles and start to decay at $q \sim 10^3$ cm$^{-1}$ towards values that are approximately three decades smaller in correspondence to the largest $q$. The curve passing through the experimental data represents the theoretical prediction based on the Mie theory. It has been obtained by using the actual (certified) sample parameters (i.e., average diameter $\langle d \rangle = 40.3$ μm, relative standard deviation $\sigma(\langle d \rangle) = 5.7\%$, and refractive index at 633 nm equal to 1.588) and by taking into account the known array sensor
no systematic deviations

between the data and the theory is remarkably good, with
points located at very low and very large angles, the match-
geometry. Data have been rescaled by an arbitrary factor in
order to overlap with the theoretical curve. Except for a few
points located at very low and very large angles, the match-
ing between the data and the theory is remarkably good, with
no systematic deviations (the overall rms value is ~5%).

VI. TEST WITH POLYSTYRENE BEADS

To test the instrument under realistic experimental con-
ditions, measurements have been carried out on aqueous sus-
pensions of polystyrene beads drawn from the chemical plant
where the particle sizer has to be installed. Samples of
known particle size distribution have been obtained by siev-
ing the drawn polystyrene beads by means of calibrated
sieves (made by ENDUCOTT’S Ltd.). The narrowest distri-
butions obtainable in this way have widths which are about
12% of their mean diameters. Such a level of polydispersity
is small when compared with the width of the classes used to
retrieve the particle size distribution, therefore, they can be
considered almost monodisperse distributions and we will
refer to them as monodisperse samples.

Our tests were carried out both on monodisperse and
moderately polydisperse samples with diameters in the range
~300 µm to ~2 mm. The beam attenuation, set by properly
controlling the sample concentration, was varied over a fairly
wide range of values, i.e., from ~5% to ~60%.

In Table I we show the results relevant to three different
monodisperse samples. The table reports, for each sample,
the mesh size of both the low-pass and the high-pass sieves,
the mean diameter of the recovered distribution and its full
width half maximum (FWHM) width, expresses both as per-
centage of the mean diameter and as number of particle
classes. As one can notice, in all the three cases, the recov-
ered mean diameter falls within the sieved sample size range
and its accuracy is better than 10%, a value which is more
than adequate for the characterization of the polymer growth
process. The FWHM of all the recovered distributions are
somewhat larger than expected, being approximately 30%–
40% of their average diameters. This is due to the finite
resolution of the inversion method and, consistently with the
work of Ref. 8, they correspond to about 2–3 particle
classes. The uncertainty associated to the recovered mean
diameter of sample No. 2 is a reasonable estimate of the
measurements reliability. Indeed in that case, we repeated the
measurement on 20 different samples prepared with the same
sieving procedure, and we obtained always the same mean
diameters within a 0.5% standard deviation.

An example of the results obtained in the case of mono-
disperse samples is shown in Fig. 10. The figure reports the
recovered particle size distribution (solid line) and the sample
distribution obtained with two sieves of mesh sizes
315–355 µm (dashed line). Both the distributions have been
normalized to an overall volume fraction equal to 1. The
figure shows clearly that, although the two distributions have
rather different widths, their peaks match quite nicely, and
the corresponding accuracy on the recovered average diam-
eter is better than 1%. The quality of data reconstruction can
be estimated in Fig. 11(a) where the measured intensity dis-
tribution $I(q)$ and the intensity reconstructed on the basis of
the recovered particle size distribution as that of Fig. 10 are
plotted as a function of the wave vector $q$. The agreement
between the two curves is remarkable, as it is also evidenced
by the plot of the percentage deviations reported in Fig.
11(b). As one can notice, the deviations remain very small
(less than 0.1%) for low $q$ values and start to become appre-
ciable at $q$ vectors larger than 200 cm$^{-1}$, with values always
confined within 10%. This is ultimately due to the finite reso-
lution of the method which always recovers distributions
somewhat broader than the original ones. As a consequence,
the reconstructed data are necessarily smoother than the mea-

TABLE I. Results relevant to monodisperse sieved samples.

<table>
<thead>
<tr>
<th>Low-pass mesh size (µm)</th>
<th>High-pass mesh size (µm)</th>
<th>Recovered mean diameter (µm)</th>
<th>Recovered FWHM (%)</th>
<th>Recovered FWHM (No. of classes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>315</td>
<td>355</td>
<td>336</td>
<td>45</td>
<td>3</td>
</tr>
<tr>
<td>710</td>
<td>800</td>
<td>752±0.5%</td>
<td>28</td>
<td>2</td>
</tr>
<tr>
<td>1000</td>
<td>1250</td>
<td>1240</td>
<td>29</td>
<td>2</td>
</tr>
</tbody>
</table>

FIG. 9. Intensity distribution scattered by a suspension of latex spheres with
a nominal diameter of 40.3 µm and relative $\sigma$=5.7%. Comparison between
the experimental data (open circles) and the theoretical curve (calculated by
means of the Mie theory).

FIG. 10. Measurements carried out on a monodisperse polystyrene beads
sample obtained by using two sieves of mesh size 315 and 355 µm. Com-
parison between the recovered particle size distribution (solid line) and the
sieved one (dotted line).
sured ones and their matching is poorer in the $q$ regions where the intensity changes very fast, as it happens at high $q$ values in our case.

As a further test on the monodisperse samples, we investigated the possibility of utilizing the instrument in connection with highly concentrated samples. This is of practical importance since the possibility of handling concentrated samples makes it possible to relax dilution requirements during the drawing process and hasten the monitoring procedure. Of course, at high concentrations, multiple scattering may heavily affect the measurements, therefore making particle sizing unreliable. However, due to the peculiar scattering regime the instrument has to operate (i.e., a strong diffraction regime where the light scattered by very large particles is detected at very low angles) as well as to the peculiar design of the test cell (cell thickness much smaller than the average interparticle distance) multiple scattering problems are expected not to be very critical. This was confirmed by our measurements carried out on samples of increasingly higher concentrations, spanning a range of beam attenuations from $-5\%$ to $-60\%$. For all the three samples shown in Table I, we recovered particle size distributions with average diameters always equal, within a few percents, to those reported in the table. Upon increasing the sample concentration, there was only a slight tendency to recover smaller and smaller average diameters. This effect was, however, very marginal since we observed a maximum deviation of $5\%$ in correspondence to a $60\%$ beam attenuation.

Finally, we carried out measurements on weakly polydisperse samples, properly prepared so to match, as close as possible, the ideal narrowest size distributions obtainable from the growth process. Tests were aimed to investigate whether the instrument is able to appreciate a broadening of the size distribution beyond its intrinsic resolution. An example of such tests is reported in Fig. 12 where we show both the sample distribution prepared by using several couples of sieves (dashed line) and the recovered particle size distribution. As it can be noticed the matching between the two distributions is fairly satisfactory with mean diameters which differ only by less than $5\%$, being $(d) = 876\ \mu m$ for the sample distribution and $(d) = 918\ \mu m$ for the recovered one. The FWHM width of the recovered distribution is $650\ \mu m$, that corresponds to about 5 classes. Therefore, since the instrument resolution is 2–3 classes, we can conclude that differences between monodisperse and weakly polydisperse samples can be reliably appreciated. Also in this case we checked the inversion algorithm by comparing the measured and the reconstructed intensity distributions [see Figs. 13(a) and 13(b)]. As in the previously described test (see Fig. 12) the agreement between the two curves is fairly good. In this case, because of the broader particle size distribution, the matching is even better, with percentual differences always smaller than $2\%$.

**ACKNOWLEDGMENT**

The authors would like to acknowledge Professor M. Giglio of the University of Milan, Physics Department for the fundamental support in the design of the instrument.


