Direct measurement of pair correlation in model aerosols.

Peter D. Scott, Ph.D.

State University of New York at Buffalo
Capen Hall
Amherst NY 14260

U. S. Army Research Office
P. O. Box 12211
Research Triangle Park, NC 27709-2211

The feasibility of using in-line Fresnel holography, together with digital decoding algorithms which operate on the unreconstructed hologram, to estimate the pair correlation function of model particle fields simulating aerosols and hydrosols at smaller scale was investigated. New algorithms for digital decoding which incorporate super-resolution signal processing were developed and demonstrated. This method was judged a useful adjunct to theoretically based state truncations, pure digital simulations and x-ray structure factor measurement for estimation of pair correlation.

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DIRECT MEASUREMENT OF PAIR CORRELATION IN MODEL AEROSOLS

PETER D. SCOTT, Ph.D.

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INTRODUCTION

This Final Report summarizes the principal results of research activities undertaken under Army Research Office Grant/Contract No. DAAL03-89-K-0134, 1 July 1989 - 30 September 1990, under the auspices of the Research Foundation of the
State University of New York. Additional, more detailed material is included in the manuscripts cited in the Bibliography as entries [1]-[8], each derived in part from the support and activities of this contract.

The Principal Investigator Peter D. Scott, Department of Electrical and Computer Engineering (ECE), State University of New York at Buffalo (SUHYB), was aided in this project by several able individuals. Mr. Lifan Hua, candidate for the Ph.D. degree in ECE SUNYB, assisted in all aspects of the work and was supported under the contract for a brief period of time. It is anticipated that he will complete requirements for the Ph.D. degree within the next six months, partly as a result of his activities and support under this contract. Mr. Gongweil Xie, ECE SUNYB Research Associate, contributed extensive efforts at the optical bench without any charge to the contract. Dr. David T. Shaw, Professor of ECE SUNYB, offered his aerosol laboratory facilities and supplies, along with frequent technical consultations and suggestions, also without any charge to the contract. Without the magnanimous support of these two individuals, scope and activities would have been much attenuated.

As more completely described in ARO Proposal Number 27078-GS, the central focus of the research activities supported under this contract was investigation of the feasibility of utilizing Fresnel holography, together with digital reconstruction techniques, to assess the pair correlation of model particle systems. Experimental activity consisted in the fabrication of an optical cell in which the particles could be suspended and of the particle systems themselves, alignment of the YAG laser and optical bench for Inline Fresnel holography, and acquisition and processing of the holograms of the optical cell contents. Representative sections of the processed holograms were subsequently digitized under high magnification, input to a general purpose computer, and analyzed using
algorithms we developed to estimate each visible particle's centroid location at the highest attainable resolution. The resulting three dimensional data base of particle location parameters was mapped into an pair correlation function estimate using standard non-parametric distribution function estimation technique.

BACKGROUND

By identifying interparticle separations of higher and lower probability than the completely random case, the pair correlation function (or, in the isotropic case the radial distribution function) reveals preferred spacing intervals even in materials such as gases and fluids that have no obvious long range order. These preferred spatial scales predict wavelength dependent scattering and attenuation effects for wave structured energy propagating through the particle field. Anisotropic pair correlation suggests dependence on the direction of wave propagation relative to the orientation distribution of the particle ensemble as well as wavelength dependence.

The pair correlation is a primary input parameter in recent predictive multiple scattering theories of wave propagation through aerosols and hydrosols. In the Varadan approach [9]-[11], the quasicrystalline approximation is used to characterize ensemble field coefficients in terms of the pair correlation, from which a dispersion equation is derived using Twersky's extinction theorem. This method for prediction of scattering and attenuation of wave energy is of broad generality, applying to both electromagnetic and compression waves in air, water, vacuum and other media.

Techniques employed to estimate the pair correlation of "interesting"
particle fields include truncations based on theoretical arguments (such as Perkus–Yevich and hypernetted chains), digital computer simulation employing Monte Carlo or molecular dynamics, and measurement of x-ray structure factor. Truncations of the equations of state yield systems which can only be solved, or even reliably estimated, in the simplest cases such as monodisperse dilute hard spheres. Lagos and Seville [12] have recently solved the Perkus–Yevich equation numerically for hard spherocylinders (particles formed by capping right circular cylinders with half-spheres). They failed to find even rough corroboration for current pair correlation mixture estimates based on perturbation theories or simplified integrals.

Digital simulations are much more general in scope and model parameters, but for realistic polydisperse dense particle fields, simplifying assumptions must be applied which bring the results into question, and even then computing time becomes a cost problem even on modern high speed digital computers. This is particularly evident in the case of relatively high density non-spherical particles [13]. X-ray structure factor measurement is an attractive, direct method for pair correlation estimation, but is limited to cases where particles are of the same granularity as the x-ray wavelength. Thus the need exits for a method of assessment of pair correlation which would complement the capabilities of the existing techniques. This is the opportunity addressed by the holographic method for pair correlation measurement in model systems investigated in the present research project.

PROBLEM STATEMENT

The central problem to be addressed is the accurate assessment of the
distribution of centroids of every particle within a characteristic test volume of the optical cell. Underlying this mensuration goal is the hypothesis that the ensemble of model particles, scaled in size relative to the target particle field whose pair correlation is to be estimated, will accurately reproduce the (scaled) pair distribution function of the target particle field whose pair correlation is being estimated. Note that this assumption is plausible only in the non-interacting hard particles case (in which the only interparticle interaction consists of a non-interpenetrability constraint). Other particle-particle interactions, such as electrostatic or magnetic coupling, would be more difficult to simulate in the model particle field with accuracy, since in general such forces do not scale coherently with particle dimension.

The three dimensional particle centroid location database must be acquired within a few seconds after the model particle field is last stirred, in order to prevent settling and distortion of the particle separation and orientation geometry. Each particle in a sample portion of the optical cell must be observed and its centroid computed accurately. The accuracy of centroid location, i.e. the resolution of the measurement system, must be significantly greater than a standard particle length in order to detect and locate narrow features (peaks and nulls) in the pair correlation function with useful accuracy. Finally, the resulting list of particle locations in three space must be mapped into a pair correlation estimate, taking into account the resolution-smoothness tradeoff inherent in all probability density function estimation methods. The estimation method must have sufficient resolution to detect significant fluctuations from the purely random distribution case even when they are relatively narrow, while preserving as low an estimate variance as is attainable consistent with the required resolution.
METHODS

The optical cell used was approximately 2 cm on a side, made of high quality optical glass, with opposing surfaces carefully aligned and glued for parallelinity. The particles were standard latex spheres of various sizes ranging from 5 micrometers to 100 micrometers, as well as cylindrical iron fibers produced in our laboratory, microtomed and sieved to size. The media in which the particles were suspended was high purity water. All experiments were done at room temperature.

The optical setups are shown in Figures 1 and 2. Figure 1 shows the in-line hologram acquisition configuration, in which the optical elements are illuminated by a YAG laser. A high power pulsed laser was utilized to permit shortening exposure time sufficiently so that high resolution, relatively insensitive hologram emulsions could be used and yet avoid motion blur. The optical cell was located at position D. Figure 2 shows the monitoring configuration used to check digital reconstructions by direct optical measurement. The conventionally reconstructed image is routed to to TV monitor and via analog to digital conversion (ADC) to a general purpose computer for comparison with digital reconstruction (DEC VAX 780 and SUN 3/260 computers were used). The monitor was useful in surveying the holograms, tuning the digital algorithms and verifying their basic particle counts along with coarse verification of the centroid computations.

The digital algorithms used to analyze the contents of the RD converted holograms are based on the twin-image elimination methods previously developed in this laboratory [14]-[15]. In order to maximize the resolution (thus reducing the
possibility of missing closely opposed particles and reducing the centroid location error variance of the ensemble) a "superresolution" signal processing layer was built upon this base [1], [2]. In this technique the bandwidth limitation of the reconstruction effectively imposed by the physical size (hologram plate format) limitation is stretched by an iterative spectral extrapolation procedure. The effect is to computationally recreate a portion of the hologram that was lost at the borders of the holographic plate thereby permitting higher frequency components (i.e. higher resolution) in the object field reconstruction. This algorithm is designated as the Phase Retrieval Spectral Continuation (PRSC) algorithm. Finally, the mapping from detected centroid locations in three space to a final pair correlation function estimate was made using the standard Parzen function method.

RESULTS

A central result of this study was the successful upgrading of algorithms originally designed for general particle field inline hologram decoding to the present purposes. The resolution of the algorithms presented in [14] and [15] needed to be improved to permit sufficiently accurate and consistent determination of the centroid locations of each of the particles in the holographic test region.

The referenced algorithms effectively reproduced by digital means the resolution available with high quality conventional (optical) resolution once the twin image artifact had been suppressed. This required enhancement for effective pair correlation estimation for two reasons. First, the reduced axial resolution inherent in in-line holography [16] together with the shadowing of particles
located in a narrow cone behind more proximal particles (relative to boresight distance from the laser source) made it possible that deeply shadowed particles might not be detected. This effect must be minimized, since it leads to undercounting of closely opposed particles which happen to be aligned along the optical axis, and thus to systematic bias reducing the pair correlation at low separations. Second, the error variance of the pair correlation estimate at given separation is directly proportional, in a given run, to the individual variance in the estimate of centroid-centroid separation for a given particle pair falling within that measurement bin reduced by the square root of the number of particle pair separations within that bin. Since there are relatively few particle pairs at the smaller separations, the variance is highest exactly where the pair correlation is of most interest and must be reduced as much as possible in order that interesting features not be lost under the measurement noise.

This enhancement was achieved by using superresolution signal processing methods described in [1] and [2]. An increase of factors of two to eight was measured in the available resolution, dependent on experimental signal to noise ratio. To determine the likelihood of missed particles, simulations were employed. It was found that only under very low probability circumstances, such as the complete shadowing of a smaller particle within one to two interparticle separations directly behind the larger one was a particle likely to be missed. Thus at all particle densities consistent with accurate in-line measurement, i.e. at least 75% of the illumination reaching the hologram along the undiffracted reference pathway [16], the resolution of the resulting technique was satisfactory to the present purpose. Studies of both latex spheres and cylindrical particle populations at low densities (up to 0.05% volume fraction density) indeed verified this assertion. Within experimental error the resulting
pair correlation estimates had the familiar "well stirred" configurations.

Unfortunately, the optical cells available for this project proved unsuitable for higher density trials. In order to preserve the 75% criterion, a straightforward calculation shows that at the volume fraction of 1%, the border of interestingly "turgid" fields, it is necessary that the cell be only on the order of ten particle lengths thick (measured along the optical axis; the other dimensions may be arbitrarily large). The narrowest cell available was 2 cm thick, requiring particles too large to be effectively hologrammed. Our efforts to fabricate cells with thickness 1-2 mm were unsuccessful within the time and cost constraints (the fabrication effort was unsupported in this contract). Difficulties included the very narrow separation distances and the high degree of parallelinity required to prevent coherent reflection artifact.

CONCLUSIONS

The principal conclusion of this feasibility study is that in-line holography, supported by digital reconstruction and resolution enhancement, is a viable complement to analytical approximations, digital simulations and x-ray structure factor measurement for the determination of pair correlation within some model aerosol systems. At low densities the well stirred solution is recovered within experimental error. At higher densities very narrow optical cells are required to maintain the quality of the in-line holograms. While no experimental data was available, all-digital simulations suggest the superresolution algorithms should be equally applicable in the high density as in the low density regime.
BIBLIOGRAPHY


**Figures**

**Figure 1.** Hologram acquisition optical bench setup.

**Figure 2.** Optical reconstruction and monitoring.
APPENDIX

The first pages of manuscripts [1]-[8] listed in the Bibliography follow in order. Each of these manuscripts was based in part on the activities and support of ARO Grant/Contract DAAL03-89-K-0134. The brief duration of the contract (12 months with a three month extension) explains why most of these manuscripts are under review, or accepted but not yet appeared, at the time of closure of this Final Report.
An adaptive constraint algorithm for image understanding of digitally reconstructed in-line holograms
by
Lifan Hua, David T. Shaw and Peter D. Scott
Dept. of Electrical and Computer Engineering
State University of New York at Buffalo
Bell Hall, Amherst NY 14260

Abstract

Accurate quantitative decoding of the 3D information encoded in in-line holograms, at best difficult optically, may be achieved by A/D sampling of the hologram followed by digital reconstruction. Using standard reconstruction technique, the resulting image bandwidth is limited by the size of the hologram. In addition, the phase ambiguity inherent in magnitude-only hologram recording yields, along with the desired object reconstruction, an out-of-focus conjugate artifact called the twin image. Both limited bandwidth and twin image restrict the available resolution well below the theoretical diffraction limit. In this paper an algorithm is presented which addresses both problems by iteratively combining phase retrieval and spectrum continuation to produce estimates of the phase of the recorded hologram, and both magnitude and phase of the hologram beyond its physically recorded boundaries.

Since algorithms based on spectral continuation are sensitive to the constraints imposed on the extent of the objects being imaged (which must be space limited), a method for selecting these constraints adaptively was developed. Effective use of adaptive constraints greatly accelerates convergence of the algorithm compared to fixed constraints. The chosen adaptive constraint selection rule is shown to have an error-correcting property in which over-constraint is naturally corrected and the constraint set boundaries stably relax toward the actual object boundaries. Examples demonstrate the degree of improvement in resolution over conventional reconstruction, and over reconstruction with phase retrieval only. This algorithm suggests the possibility of super-resolution holography, in which the diffraction limit is exceeded. While difficult to achieve in practice, super-resolution has been demonstrated for other kinds of imaging but not heretofore considered for holography.

1. Introduction

Under a variety of conditions a multidimensional signal may be reconstructed from partial information about its transform, for instance its Fourier or Fresnel transform[1-4]. While the ability to reconstruct a signal from magnitude information only would be useful in several applications settings,[5-9] magnitude alone is not in general sufficient to reconstruct a signal. For example, holography is a useful modality for capturing an object set's three dimensional distribution. But only the intensity of the optical field is recorded on the film emulsion or other photosensitive medium.

The clarity and resolution of conventionally reconstructed holographic images is limited by factors including size of the hologram plate, grain and the nonlinearity of the holographic film and the twin image artifacts. Techniques for suppressing twin image in the digitally reconstructed in-line holography by phase retrieval and by modified inverse filtering have been reported [5,6]. These techniques do not seek to overcome the reconstruction bandlimitation inherent in the finite hologram size. In the case where missing high frequency components is the dominant resolution restriction, the resolution of the reconstructed images will not be improved much no matter how precise the estimated phase information is. In this paper, a new method for "continuing" or extrapolating a given hologram is described. This method employs a convolution/deconvolution iterative continuation cycle, combined with interior phase retrieval since the missing phase information is required for successful continuation. This algorithm uses a Gerchberg-Saxton like "error energy" reduction principle.

With standard constraint specifications, considerable computational time is needed due to the slow convergence of the combined phase retrieval and spectrum extrapolation (phase retrieval and spectrum continuation are each known to converge slowly; here both are being solved simultaneously). By adaptive selection of the constraints in the object domain, the convergence may be greatly accelerated without inducing instabilities or errors.

2. Background and problem formulation

It is assumed in this paper that the objects are opaque and the background is transparent, and the thickness of each individual object can be ignored. Thus the objects being imaged can be considered as a set of (cross-sectional) planar distribution function in the 3D object space. These assumptions are motivated by the study of particle fields
Resolution Enhancement in Digital In-line Holography

Lifan Hua, G. Xie, David Shaw and Peter Scott
Department of Electrical and Computer Engineering
State University of New York at Buffalo
Buffalo, NY 14260

Abstract:

The image volume captured in a in-line hologram may be reconstructed optically or, following analog-to digital conversion, algorithmically with the aid of a digital computer. Factors limiting the clarity and resolution of the resulting reconstructions (both optical and digital) include size of the hologram, grain and nonlinearity of the film, and twin image artifacts. Applications emphasizing display and visualization require optical reconstruction, while technical applications demanding detailed quantitative image volume descriptions favor digital technique.

An added advantage of digital reconstruction is the ease with which postprocessing may be applied to suppress noise and artifact, or enhance resolution. In this communication a constrained iterative “super-resolution” type algorithm is described and applied to enhance resolution of in-line holograms of the particle field type. This algorithm is based on the unique relationship between a space-limited object and any finite segment of its spectrum. If the truncated complex spectrum of a space-limited object is measured or derived, the full spectrum may be continued in only one way.

For in-line holograms, only the magnitude of the holographic field is recorded. To achieve super-resolution, both magnitude and phase information are needed for spectrum extrapolation. Therefore, a phase retrieval scheme is joined with spectrum extrapolation in the present algorithm. This method uses a Gerchberg-Saxton like convolution/deconvolution iterative cycle to trim the iteratively estimated data subject to the constraints of the object domain and hologram domain. The constraint in the object domain is that the object has finite size in space, and in the hologram domain the constraint is the known measured magnitude.

The super-resolution algorithm has been tested on both computer generated holograms and real optical holograms. Results demonstrate the ability to significantly improve resolution while suppressing the twin image. the algorithm is shown to be robust against noisy data. The algorithm permits accurate and high resolved quantification of dynamic 3D particle fields.
In this paper, an optical technique which uses divergent beam, high magnification microscope objective lens and double exposure method to record holograms of fast moving fibers (125 meters/second) with submicron diameters (0.5 microns) is reported for the first time. The principle of using divergent laser beam to protect high magnification microscope objective lens based on ray-tracing equations and the calculation of magnification of whole process for recording and reconstructing holograms based on optical theory are presented. An in-line double exposure holographic experimental system, utilizing the principle of dividing one beam to two parts with different optical distance, was set up to record the holograms of fast moving fibers. A set of optical reconstructions of the holograms are given and the measuring results are presented. The results show that by using the method above mentioned, the accurately measuring results of both fiber’s submicron diameter and high velocity can be determined at the same time.
Particle Measurement Using Overlapping Optical Field Technique

Gongwei Xie, Peter Scott, David T. Shaw

Department of Electrical and Computer Engineering
State University of New York at Buffalo
Buffalo, NY 14260

Yimo Zhang

Department of Precision Instrument Engineering
Tianjin University
Tianjin, P. R. China

Abstract

An optical scattering system is described which uses the Overlapping Optical Field technique (OOF) for simultaneous measurement of particle size and velocity. Experimental results using this system to study rapidly moving small particles of varying size, shape and density are presented.

Key Words: overlapped optical field, light scattering, particle measurement, polarized direction, measurement volume.
A New Optical Technique For Particle Sizing And Velocimetry

by

Gong-wei Xie, Peter Scott, David T. Shaw

Department of Electrical and Computer Engineering, State University of New York at Buffalo, Buffalo, NY 14260

Yimo Zhang

Department of Precision Instrument Engineering, Tianjin University
Tianjin, P. R. China

Abstract

A novel Overlapping Optical Field (OOF) technique that transforms an optical field of Gaussian intensity into one with uniform intensity is described. Comparison of calculated and measured results demonstrate that a highly uniform optical field in one transverse dimension with efficient utilization of optical energy can be obtained by using the OOF technique. This technique can be used for particle sizing and velocimetry.

1. Introduction

A uniform intensity distribution across a laser beam is important in particle measurements using Mie theory. Since a laser beam has a Gaussian intensity distribution, signals scattered by a small particle passing through the center of a beam and by a larger particle passing through a region of lower intensity may not be distinguishable. Several techniques have been developed to solve this problem, which can be divided into two groups. In the first group, the Gaussian distribution is directly used to infer properties of the particle size and velocity distribution [1-3]. In the second group, the Gaussian distribution is first transformed into some approximation of a uniform intensity distribution [4-6]. In this paper, a novel single-particle measuring technique of the second type, designated as the Overlapping Optical Field (OOF) technique, is described.
Using dynamic holography for iron fibers with submicron diameter and high velocity

G.W. Xie, L.F. Hua, S. Patel, P. Scott, D.T. Shaw

State University of New York at Buffalo, Department of Electrical and Computer Engineering, Buffalo, New York 14260

ABSTRACT

A divergent laser beam was used to record holograms of moving iron fibers with submicron diameter (0.5 microns) and high velocity (125 meters per second). Analysis based on geometric optics is used to derive the magnification factors for recording and reconstruction. Experimental results consisting of single-shot holograms of fast moving fibers are presented. This technique can be used for dynamic measurement of particle size and fiber orientation distribution.

1. INTRODUCTION

The in-line far-field holographic technique has been used for particle measurement for many years, and a wide variety of useful results have been obtained. But up to the present, there is no report in the literature demonstrating dynamic measurement of moving fibers particles with both submicron diameter and high velocity (over 100 m/sec).

A difficulty needing to be overcome in this case is that dynamic holography of very small, rapidly moving particles fields requires both the use of a microscope objective lens of high magnification and long working distance, and high power pulsed laser. For in-line holography, a plane wave is the obvious choice to illuminate the particle field. But as the convergent point of microscope objective lens is always close to one element of the objective lens, when the incident wavefront is planar the extreme power densities caused by focusing a high power pulsed laser introduces heating of the lens surface sufficient to damage the lens. In this paper we will present a divergent beam method which prevents damage of the microscope objective lens. The pulse energy received by the surface of the last piece of the microscope objective lens is spread over a considerably larger cross-section than the planar case. Analysis by geometric optics shows that an approximately 6° divergent beam spreads the energy over 60 times as large a surface as the corresponding planar beam, shifting the incident radiant power density from well above the amount lethal to the lens to safety below.

In Section 2 the divergent beam method is described and a quantitative analysis of the power density impinging on the microscope lens over the range of beam divergence angles from 0° to 7° is presented. In Section 3 the magnification factors for recording and reconstruction are calculated. Section 4 describes the results of divergent beam in-line holography for fast moving narrow gauge iron fibers, and includes an error analysis. A summary is offered in Section 5.

2. DIVERGENT LASER BEAM METHOD

The microscope objective lens (ML) which we used for taking dynamic holograms of fine fibers is a product of Bausch & Lamb Corp., 25x 0.31NA. It is a compound objective lens which consists of five lens pieces: ML1, ML2, ML3, ML4, and ML5. Two of these are Fraunhofer cemented doublet lens, ML1, ML2, ML3, ML4 are positive lenses and ML5 is a negative lens. The structure of ML is sketched in Fig.1.

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Single Shot Holography of Fast Moving Fibers with Submicron Diameter

G.W. Xie, L.F. Hua, S. Patel, P. Scott, D.T. Shaw

State University of New York at Buffalo, Department of Electrical and Computer Engineering, Buffalo, New York 14260

ABSTRACT

A divergent laser beam was used to record holograms of moving iron fibers with submicron diameter 0.5 μm and high velocity 125 meter per second. The principle of divergent laser beam method based on geometrical optical theory and the magnification calculation of recording and reconstructing process of hologram are presented. The experimental results of single-shot holograms of fast moving fibers are demonstrated. This technique can be used for dynamic measurement of particle size and fiber orientation distribution.

1. INTRODUCTION

The in-line far-field holographic technique has been used for particle measurement for many years and great successful achievements have been obtained in this field. But up to the present, there is no report in the literature about the dynamic measurement of moving fibers particles with both submicron diameters and high velocity (over 100 m/sec). The reason is that dynamic holograms of very small rapidly moving particles fields requires both the use of microscope objective lens of high magnification with long working distance and high power pulse laser. According to the general principle of in-line holographic technique, the plane wave is usually used to record the hologram. But as the convergent point of microscope objective lens is always close to one piece of the lenses which compose the whole objective lens when incident is plane wave, the extreme power densities derived by focusing a fast high power pulse laser introduces heating of the lens surface sufficient to destroy the lens. In this paper we will present a divergent beam method to avoid the damage of the microscope objective lens. By this method, the pulse energy received by the surface of the last piece of the microscope objective lens can be spread over a considerably larger cross section than the planar beam is used. Analysis by geometric optics shows that an approximately 60° divergent beam covers 60 times as large a cross section of the entire lens surface as the
IMPORTANT MESSAGE

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