Phase in Nanooptics

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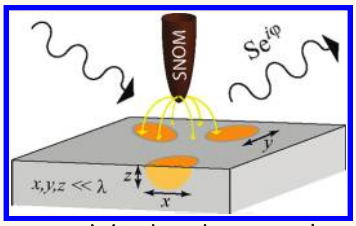
ABSTRACT



Quantitative phase measurements in imaging, microscopy, and nanooptics provide information not carried in amplitude measurements alone. In this issue of *ACS Nano*, Honigstein *et al*. present a new method in phase measurement. In this Perspective, we comment on this work and more broadly on the emerging role of phase and phase measurements in nanooptics.

Phase has played important roles in modern physics, and more specifically in microscopy. The concept of phase is due to Fourier and the introduction of harmonic analysis almost 200 years ago. While it may seem straightforward, our understanding of phase continues to progress on both fundamental[1] and technological fronts. Phase is central to a complete understanding of any electromagnetic or optical system. After all, Maxwell's equations do not govern the behavior of intensities, but rather electromagnetic fields, both amplitude and phase.

Measurement of optical phase once revolutionized classical microscopy. The phase contrast microscope of Zernike[2] and its cousin the Nemarski microscope[3] made visible weakly absorbing structures, such as those commonly encountered in cell biology. Because phase contrast is sensitive to small changes in optical path length, taken together with a prior assumption that rays simply propagate along straight lines through the sample, it can provide images sensitive to nanoscale changes in sample optical thickness (see Figure 1a).

Without prior knowledge, however, far-field microscopes cannot probe structures smaller than roughly half the wavelength of light. In the visible spectrum, lateral spatial resolution is thus limited to hundreds of nanometers, and in the mid-infrared to several microns. This problem can be overcome by nanooptical imaging techniques such as scanning near-field optical microscopy (SNOM), which provides insight into the structure of samples on deeply subwavelength scales. A particular kind of SNOM utilizes light scattered at a sharp probe (often an AFM tip) placed in the near-field of a sample surface (scattering-type SNOM, s-SNOM).[4] It has become a powerful tool for nanoscale optical imaging and spectroscopy due to its wavelength-independent resolution of ~10 nm at visible, IR, and THz frequencies.[4,5]

In this issue of *ACS Nano*, Honigstein *et al.*[6] present a new method of quantitative phase measurement in optical microscopy. They bring the tools of nanooptical imaging, namely SNOM, to bear on interferometric detection of phase in the Fourier plane of a sample. In this Perspective, we comment on the emerging role of phase in nanooptics.

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The measurement of phase is always, to some degree, inferred. Detectors measure power rather than fields, so relative phases between fields are often inferred from the effect of the phase on interference patterns. The interference of electromagnetic waves forms the basis of many well-known physical phenomena, including double-slit diffraction patterns, laser speckle, and holographic imaging. Those experiments in which a reference field is controlled to infer the phase are usually called interferometric.

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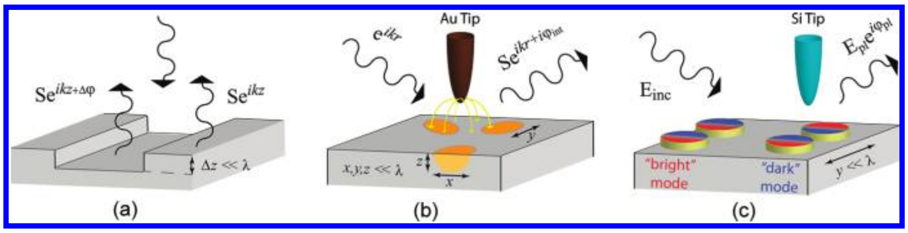

Figure 1. Phase plays three important roles in nanooptics. (a) Far-field determination of nanoscale differences in surface height, or, alternatively, sample thickness may be accomplished through phase-sensitive measurements. The phase varies over a 2π range. (b) Near-field investigation of material properties. Local interactions between the metal tip and the sample, which provide strong field concentration, change the phase of the scattered field allowing local identification of materials at nanoscale. (c) Near-field mapping of surface modes. Mapping phase of the local field distribution enables direct investigation of the modes in plasmonic nanostructures. The role of the tip is to out-couple the local field to the detector, therefore dielectric tips, which minimize the field disturbance due to the tip presence, are preferred.[28]

When applied to phase-sensitive imaging or microscopy, the chief challenge of interferometric methods lies in determining the unknown amplitude and phase profiles uniquely. Heterodyne interferometry is one such strategy in which the frequency of the reference beam is shifted slightly in order to produce a temporal beating of the detected intensity well within the bandwidth of optical detectors. The phase of the beat frequency is dictated by the relative phases of the signal and reference beams, and it can be easily measured by comparing it to a reference at the same frequency with constant phase. Historically, heterodyning has been the technique of choice for phase-sensitive monochromatic SNOM.[4,7−12]

As an alternative to heterodyning, multiple intensity measurements may be made with distinct, known reference phases. As few as two measurements are sufficient to determine the complex signal field, although more measurements improve performance. So-called phase-shifting interferometry has long been used in the optical testing community, has been implemented in SNOM, and has recently been implemented in near-field microscopy to visualize the complex near-field coupling tensor between two gold nanoparticles.[13] Phase-shifting is a time-domain solution to the phase-resolved interferometry problem, which enables relative simplicity in near-field experiments. Since the principles are well understood in optical testing, error analysis has the potential to be sophisticated in nature. Fourier-transform interferometry can be seen as a broadband extension of phase-shifting interferometry, in which phase shifts at many wavelengths are imposed simultaneously. This technique enables the mapping of IR spectra with nanoscale spatial resolution, which is an improvement of several orders of magnitude compared to conventional far-field infrared spectroscopy.[14]

Between heterodyne and phase-shifting interferometry lies phase-modulation, or pseudoheterodyne interferometry. The reference phase is quickly and continuously modulated, producing a dynamic frequency shift. The resulting interferometric intensity contains amplitude and phase information encoded at alternating harmonics of the modulation frequency. Pseudoheterodyning has met with success in the SNOM community in applications of material and free-carrier mapping[5] and near-field visualization of optical antenna modes.[15]

If only propagating optical fields are considered, there is one-to-one mapping between an object and its spatial Fourier transform. In principle, it does not matter which of the two is measured, the same information is obtained. Interferometry therefore has a natural extension to the spatial-frequency domain. This idea is the basis for the high-gain avalanche rushing amorphous photoconductor system (HARPS), the heavy-atom X-ray diffraction technique after which the technique of Honigstein *et al*. is modeled, as well as other phase-resolved imaging systems. In these techniques, a point-like light source is placed in or near the object plane. In the Fourier plane, far from the object, the point-like source appears approximately as a plane wave, which interferes with the Fourier transform of the image field to create interferometric intensity. The technique also bears a resemblance to point-diffraction interferometry.[16] In the paper of Honigstein *et al*., the point-like object is the sharpened end-facet of an optical fiber and the sample of interest is microscopic.[6] They obtain phase-resolved images of two test samples, demonstrating the lateral and axial resolution necessary to image a fresh red blood cell. Although the near-field of the sample is not probed, the authors point out that by acquisition of multiple images and data processing, the resolution of the final images could be improved, perhaps beyond the usual diffraction limit.

Interferometry is not the only phase inference technique. Noninterferometric determination of phase presents a daunting mathematical and computational challenge. It is nonetheless necessary in many situations in which a coherent reference is not practically available. This is the problem that astronomers face, for example, because they have no control over distant galaxies. Phase-retrieval algorithms have been developed for cases like this, in which object phase profiles are iteratively improved from an initial guess until some termination condition is met.[17] To our knowledge, such phase-retrieval algorithms have not yet been applied to nano-optics, leaving open a potentially interesting avenue of research. Constraints on sample size and discrete constituents could prove especially powerful in the near-field.

In microscopy and nanooptics, phase variations (contrast) in images can be categorized into three fundamentally distinct phenomena: (i) field propagation between the sample and the detector (propagation phase), (ii) phase-induced variations due to local optical near-field interaction between the probe and the sample (interaction phase), and (iii) phase-associated variations with the field distribution in the sample near-zone (local field phase). These phenomena are illustrated in panels a, b, and c of Figure 1, respectively.

The propagation phase is related to the optical path length experienced by free-propagating fields and manifests changes in the refractive index or physical height of a sample. This effect underpins the phase-contrast microscope. Importantly, when the phase-contrast image can be assumed to map to the real object structure, sample thickness may be determined with nanometer precision directly from the measurements of (propagation) phase (see Figure 1a). This has given impetus to the pursuit of quantitative phase measurements in far-field microscopy.[18,19]

The local interactions between the near-field probe and the sample, which is absorbing and scatters the incident field, give rise to the interaction phases (see Figure 1b). Because the interactions take place over distances much shorter than a wavelength, the propagation phase may often be safely neglected in the so-called quasistatic limit. The local properties of the sample in the vicinity of the tip are thus encoded in the field and can be imaged and analyzed. Interactions between a sample and a probe result in a phase shift of the scattered light, and this interaction is strongly material dependent. Therefore, different materials exhibit different phase fingerprints in such a nanooptical image. As an elegant example, it can be shown that if the probe–sample interaction is nonresonant, the interaction phase is simply proportional to the absorption in the neighborhood of the tip. So, performing a SNOM experiment across a wide range of frequencies can yield absorption spectroscopic information with nanoscale resolution.[20−22]

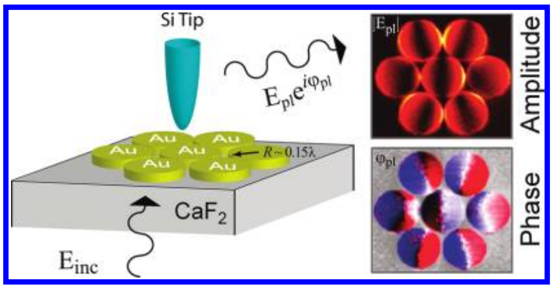

Figure 2. Principle of near-field imaging of a plasmonic structure (here a mid-infrared-resonant oligomer structure composed of seven gold disks) by s-SNOM (left). The light scattered by the tip is detected by an amplitude- and phase-resolving interferometer, yielding near-field amplitude and phase images (right). While the amplitude image shows the dipolar plasmon modes on each disk, only the phase image reveals the relative phase shift between them. The central disk oscillates out of phase with the outer disks, revealing the subradiant ("dark") character of the mode.

Finally, phase images can represent the local phases of the electromagnetic fields in the near-sample zone, thereby enabling direct mapping of optical modes supported by the sample. Among these are surface plasmon polaritons—a coupled oscillation of light and free electrons in the sample, which are central to the emerging field of plasmonics (Figure 1c). In plasmonics, metal structures often act as optical antennas, converting propagating incident optical radiation into nanoscale-localized energy of plasmon oscillation. Because plasmon modes arise from a resonance phenomenon, their phase is associated with the illumination frequency, spanning $\pi/2$ radians as the resonance is crossed. This suggests that the phase of a local near-field can be controlled, adjusted, and manipulated by an appropriate choice of frequency and polarization of the incident illumination. Applications of this phase shift include (ultrafast) coherent-control applications[23] and sensing-based applications of Fano resonances in plasmonic systems.[24] Recording both amplitude and phase of the near-field distribution enables the identification and verification of plasmon modes in basic antenna structures.[25−27] As an example, Figure 2 shows a phase-resolved s-SNOM image of an oligomer designed to produce a Fano resonance.[28] The phase image unambiguously indicates the presence of a subradiant dark mode, which causes a Fano-type resonant spectral response of the nanostructure. The identification of this mode without phase imaging would not be possible. This concept generalizes to other plasmonic phenomena including surface guiding.

Conclusions and Outlook. Quantitative phase measurements in the near- and far-field are changing imaging science in fundamental ways. Optical imaging and inspection was once the science of lenses, mirrors, and sources. In the computer age, it is becoming a science of electromagnetics, interferometry, and computation. As Honigstein *et al.*[6] predict, "A more challenging direction will be to image 'slices' of a thick sample and piece them together, to perform 3D phase imaging." The recent demonstration of just such a technique[29] proves the power of tomographic extensions of phase imaging. A measurement of the phase is only the beginning of an investigation of an object. Phase is a property of the field rather than the object under investigation. The signal cannot always be simply related to one of the three types of phases highlighted above. In a general setting, where propagation, local probe interaction, and scattering each play a significant role, the connection between three-dimensional (3D) composition and structure, and the amplitude and phase of the measured signal are nontrivial[30] and can only be unraveled if one can "perform tomography,"[31] *i.e.*, solve the inverse problem.

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It is thus necessary to ask, how are the measurements related to the structure and composition of the sample? In the simplest scenario depicted in Figure 1a, the far-field propagation phase directly reflects sample thickness or distance to reflection (range-finding). In contrast to this, in SNOM, many scattering events within and between the probe and the sample contribute to the total scattered field. Only in special cases can the images be related to the sample properties (or field distribution) in a straightforward way. Generally, Maxwell's equations connect the fields, the sample, and the probe in a manner that enables prediction of the measurements and, given enough data, calculation of sample properties from data.

In SNOM, the solution of the inverse problem and implementation are yet to be fully realized. Two-dimensional near-field back-propagation based on interferometric measurements of near-field phase and amplitude has been demonstrated,[32] but a fully tomographic experiment remains elusive. Progress in this direction is hindered by two main obstacles. First, the near-field inverse scattering problem is ill-posed, resembling the structure of an inverse Laplace transform.[33] The evanescent nature of the near-field makes reconstructions in near-field inverse scattering exponentially sensitive to noise. Second, the solution of near-field tomography requires ways of generating multiple independent data sets. As Honigstein *et al.*[6] allude, sample tilting approaches have revolutionized 3D reconstructions in electron microscopy, but such methods are daunting in the near-field.

Practical solutions of the near-field inverse problem will need to take advantage of the well-established nontomographic techniques and to extend them. A method has been developed in the microwave regime that utilizes multiple antennas to obtain linearly independent data from which near-field tomograms can be computed.[34] Recently, a means to utilize broadband SNOM data was put forward.[35] In another proposal, the usual two-dimensional scan in SNOM has been extended to a phase- and amplitude-sensitive scan of the volume above the sample.[36]

Phase-sensitive measurements have had tremendous impact in classical far-field microscopy. Today, with the availability of cheap, massively parallel computing, the computational formation of images not achievable by classical means is made possible by quantitative phase measurements. Nanooptics is just catching up. Several promising new technologies for phase measurements, including the work of Honigstein *et al.* in this issue of *ACS Nano*,[6] are providing data for a new era of physics-based imaging and computed imaging.

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