Confocal microscopy of fluids under static pressure

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Abstract. There are few reliable methods for obtaining equations of state for fluids under static pressure. We are developing confocal microscopy to investigate fluids in a diamond-anvil cell. Unlike conventional optical microscopy, confocal microscopes collect data point-by-point, enabling three-dimensional image reconstruction. By combining these images with Fabry-Perot interference measurements, we determine the volume and refractive index, as a function of pressure, in the same experiment.

1. Introduction
Diamond anvil cells (DACs), which combine high strength with optical transparency [1, 2], have been used successfully in a broad range of high-pressure studies [3]. Synchrotron x-ray diffraction (XRD) is the “gold standard” for equation of state (EOS) measurements of crystalline solids. X-ray scattering experiments can also yield pair distribution functions and information about medium-range order for noncrystalline materials such as water [4], silica glass [5], and amorphous red phosphorus [6]. However, XRD is not well suited for determining accurate EOS for such substances. The primary goal of this work is to develop confocal microscopy to the point where we can obtain high-pressure EOS data for noncrystalline materials, such as fluids and polymers, which rival the accuracy of those obtained by synchrotron XRD measurements performed on crystalline samples. A secondary goal is to measure the refractive index \( n \) at high pressures.

Several techniques using visible light have been demonstrated to measure the EOS and/or refractive index for materials under pressure. Classical (non-impulsive) Brillouin spectroscopy allows one to measure the acoustic properties of a sample in a DAC, which can then be used to construct pressure-volume isotherms. In the backscattering geometry, knowledge of the index \( n \) is required. Brillouin spectroscopy with the forward scattering geometry does not require an index determination and has been used determine acoustic constants of polymers with reported experimental errors of 4% [7]. Impulsive stimulated light scattering [8, 9] can determine acoustic properties under pressure [10]. Optical interference methods have been used to measure the EOS and refractive index for fluids and solids under pressure [11, 12]. These methods rely on the fact that the two diamond faces in a DAC form a Fabry-Perot cavity. Transmitted or reflected light give rise to interference fringes that are analyzed to obtain the index and/or thickness of the sample [13].

In our studies, we have used confocal microscopy to determine the thickness \( d \) and refractive index \( n \) of a fluid in a DAC [14]. The principle of confocal microscopy is shown in figure 1. When the light is focused on an interface (e.g., diamond/sample), the reflected light passes through a pinhole and is detected by a photomultiplier tube (PMT). However, when the focal point is away from the interface, the pinhole blocks most of the reflected light. This effect results in peaks in the reflected
intensity, corresponding to each interface. From the peak positions, we obtain the sample thickness divided by the refractive index, \(d/n\). Fabry-Perot interference measurements are then performed to determine \(d \cdot n\). From these two quantities, we determine \(d\) and \(n\) separately. The area of the sample (gasket hole) is measured from the scanned 2D image. In this way, we are able to measure the volume and refractive index in the same experiment.

**Figure 1.** Schematic of a confocal microscope. Light emitted from the focal plane (solid lines) is focused through a pinhole. Most of the light emitted from outside the focal plane (dashed lines) is rejected.

2. **Argon under pressure**

In previous work, Hanna and McCluskey [15] investigated the EOS and refractive index of argon up to 5 GPa. Confocal microscopy was performed with a Zeiss 510 META confocal laser scanning microscope with a 514 nm excitation wavelength and a Zeiss Plan-Neofluar 10x objective. The EOS agreed with prior results. An expression for the Clausius-Mossotti factor versus density was obtained.

The refractive index of argon is not well characterized at high pressures. As shown in figure 2, values obtained by light interference (Grimsditch et al. [16]) and Brillouin spectroscopy (Chen et al. [17]) are in disagreement. The solid line, extrapolated from the confocal microscopy studies [15], shows good agreement with the Grimsditch results.

**Figure 2.** Refractive index versus pressure for argon, from Brillouin scattering (Chen et al.), light interference (Grimsditch et al.), and confocal microscopy (Hanna & McCluskey, extrapolated for pressures > 5 GPa).
3. Confocal optical profile microscopy

As shown in figure 1, a confocal microscope uses a pinhole to reject light that does not emanate from the objective focal plane. Unfortunately, this approach eliminates photons that contain useful information about the specimen. We are developing a new type of confocal microscope in which the entire beam profile is analyzed, using an area detector instead of a pinhole and PMT. This approach is termed confocal optical profile (COP) microscopy.

A recent paper by Sánchez-Ortiga et al. [18] discussed using a CCD at the focal plane of the tube lens. There is, however, a crucial difference between their approach and the one described here. In their microscope, they selected an array of active pixels on the CCD and integrated the signal, in order to emulate a pinhole. With COP microscopy, in contrast, data from the entire CCD array are collected and modeled. Photons are collected from an area much larger than a pinhole, resulting in more information and potentially much higher accuracy.

A schematic of our prototype COP microscope is shown in figure 3. A continuous wave laser beam (Coherent Verdi, 532 nm wavelength, TEM00 single mode, 2.25 mm diameter) is focused by an objective (Zeiss LD Plan-Neofluar, 20x, \( f_1 = 10 \) mm) onto the sample, which is mounted on a manual \( xyz \) stage (Thorlabs). Reflected light is focused by a tube lens (\( f_2 = 250 \) mm) onto the CCD camera (Imaging Source DMK 21BU04, 640 X 480 pixel array, 8 bit dynamic range). Images of the reflected light were collected as a function of depth (\( z \)) by scanning the objective with a piezoelectric drive (Physik Instrumente PIFOC Z-Drive, 400 \( \mu \)m maximum travel). By analyzing the CCD image versus \( z \), we can obtain an accurate value for \( d/n \).

Fabry-Perot measurements were performed with a collimated white light source and Ocean Optics USB4000 spectrometer. The white-light spectrum contained interference fringes, which are analyzed to yield \( d/n \).

![Figure 3](image)

**Figure 3.** Schematic of a confocal optical profile (COP) microscope. Laser light is focused onto a DAC. Reflected light is imaged by the CCD camera. White light and a spectrometer are used for Fabry-Perot interference measurements.
4. Results
To test the COP microscope, we investigated water in a DAC at a pressure of 0.3 GPa. At each $z$ position of the objective, intensity versus $r$ was recorded by the CCD (figure 4). The results were simulated by a model in which Gaussian beams reflected from the air/diamond and sample/diamond interfaces. Interference fringes are clearly visible and can be modeled, although the intensities do not match perfectly. Bright spots at 108.5 and 240.0 µm correspond to reflections from sample/diamond interfaces, yielding $d/n = 131.5$ µm. Combining this with a Fabry-Perot measurement, which yielded $d \cdot n = 245.5$ µm, we obtain $d = 180$ µm and $n = 1.37$. Estimated uncertainties are ~1%. The index value is in agreement with earlier work by Vedam and Limsuwan [19].

![Figure 4](image)

Figure 4. Experimental and simulated data from a COP microscope for water under pressure.

5. Conclusions
Our work has shown that confocal microscopy is a viable method for obtaining EOS and refractive index measurements for noncrystalline materials in DACs. This tabletop technique is convenient, rapid, and accurate. The COP microscope yielded results consistent with prior work. In the future, new information may be obtained with this technique. For example, with existing confocal microscopes, a decrease in detector signal may indicate that light was emitted above or below the objective focal plane. However, the decrease can also be caused by absorption, scattering, or destructive interference. The COP microscope has the ability to separate out these different effects.

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