# Electronic Supplementary Information

**Measuring Capillary Wave Dynamics Using Differential Dynamic Microscopy**

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In Fig. S1 and S2 we show results from our DDM analysis of one of our samples. In performing the DDM analysis, we fit the image structure function to $\mathcal{D}(q, \Delta t) = A(q)[1 - \exp(-\Delta t/\tau)] + B(q)$. In the main text, we plot $\tau$ vs. $q$ for multiple samples. Here, in Fig. S1 and S2, we plot $\tau$ as well as $A$ and $B$ vs. $q$ for three movies (all containing 1000 SDIs) of the same sample.
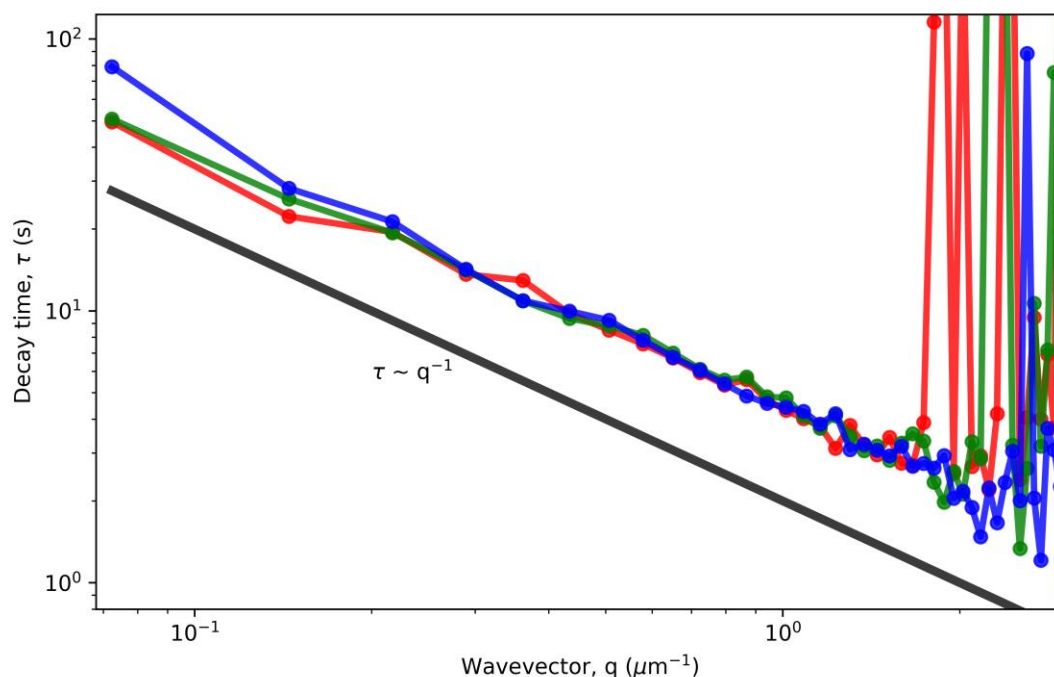


**Figure S1**. The analysis of three data sets from the same sample are shown (PNIPAM at a volume fraction of 0.1; xanthan at a concentration of 0.033% w/v). In each data set, we used a 60× 1.42 NA objective and adjusted the focus to be coincident with the liquid-gas interface. We acquired frames of 720×720 pixels (87 µm × 87 µm) at 0.32 Hz where each frame is the pixel-wise standard deviation of 50 images acquired at 42 Hz with a 10 ms exposure time. We see for all three data sets that the decay time is proportional to $q^{-1}$ which is expected for capillary wave dynamics and that the capillary velocity is about 0.46 µm/s. We observe that at around $q = 1.6$ µm$^{-1}$ the data becomes noisy. This is likely due to the fact that we are unable to accurately detect decay times less than about 3 seconds given our acquisition speed.
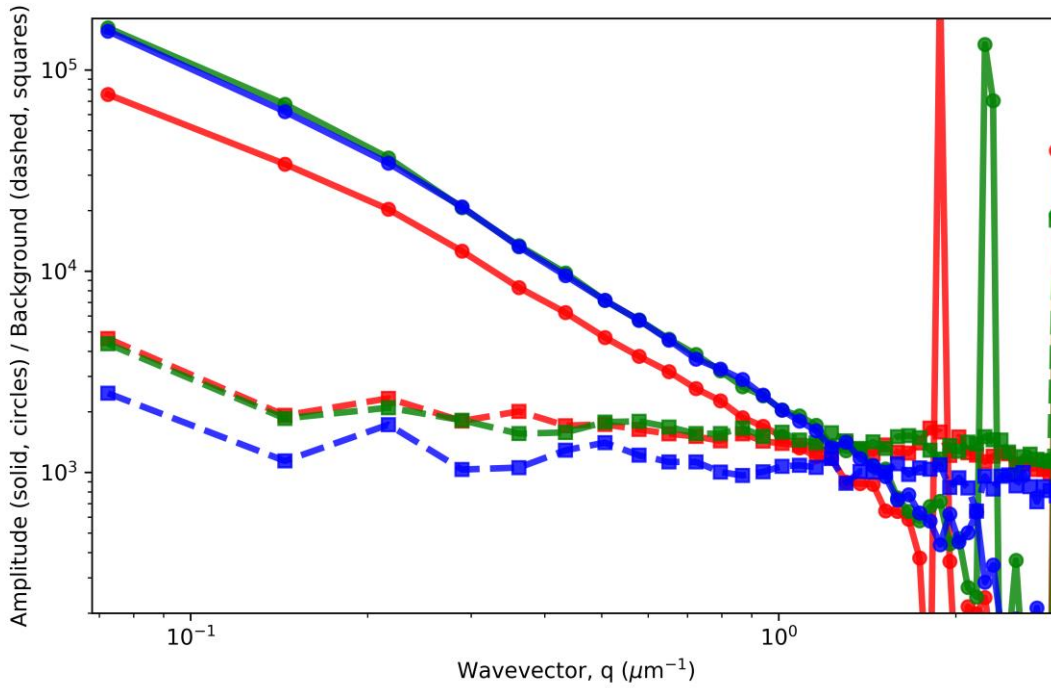
**Figure S2**. Further details of the analysis of the same three data sets featured in Fig. S1 are shown. For the DDM analysis, we fit the DDM matrix $D$ for each particular wavevector, $q$, to $D(q, \Delta t) = A(q)[1 - \exp(-\Delta t/\tau)] + B(q)$. As described in the main text, the amplitude term $A(q)$ depends on the optical properties of the microscope and the structure of the sample. The background term $B(q)$ accounts for noise. Here we show the amplitude and background parameters. The amplitude parameters are shown with the circles and solid lines and the background shown with squares and dashed lines.

In Fig. S3, we continue with data from the same sample analyzed in Fig. S1 and Fig. S2. Here, we explore the effect of not taking the standard deviation of groups of images. We see that when we perform the DDM analysis on images without taking the standard deviation, we do not observe a region where $\tau \sim q^{-1}$. We suspect that fitting our data to an alternative intermediate scattering function might reveal both ballistic and diffusive timescales which would correspond to the capillary wave dynamics and the diffusive dynamics of individual colloidal particles. However, we have not been successful in fitting our data to a double exponential. Exploring how to disentangle the dynamics when not using the SDIs for analysis is a topic of future work.
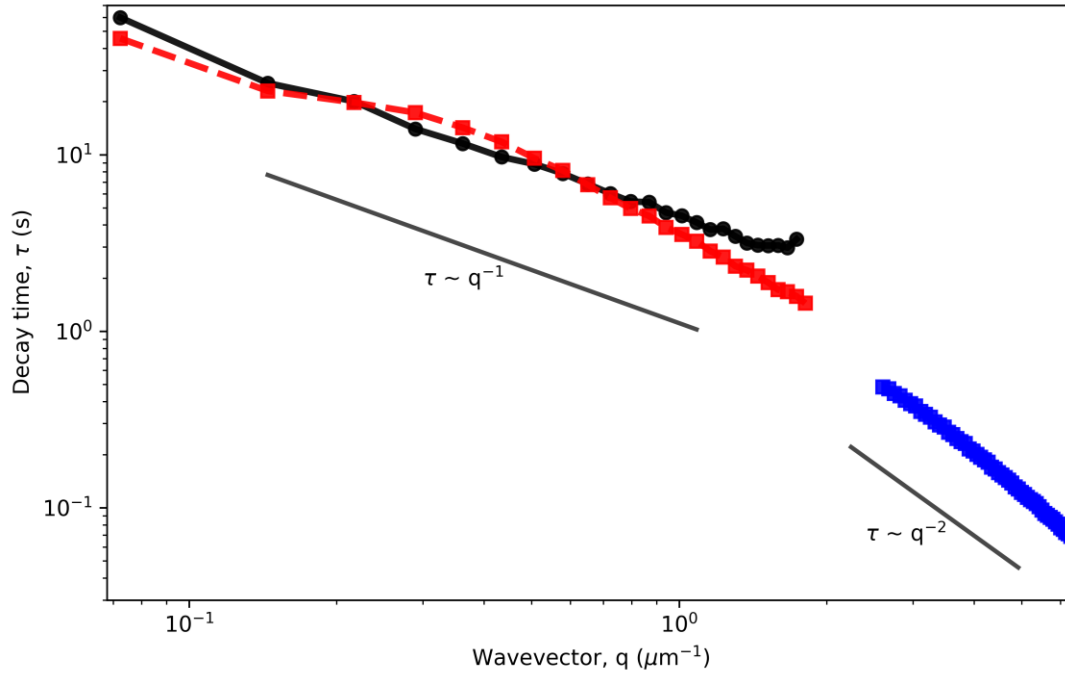
**Figure S3**. We plot the decay time, τ, versus the wavevector. Shown in black circles is the average τ of the three different data sets which were shown in Fig. S1. For each of those, we ran DDM analysis on the standard deviations (SD) of 50 images where the SDIs were acquired at 0.32 Hz. As shown, this data follows the $\tau \sim q^{-1}$ relationship. In red squares, we show the DDM analysis of images acquired at 0.6 Hz (where, as before, the images are of the planar liquid-gas interface). We use DDM to analyze the raw images, rather than the SD of a group of images. As can be seen, the red data points do not follow a consistent power law. In blue, we show the DDM analysis of images acquired at 45.5 Hz (where, as before, the images are of the planar liquid-gas interface). By taking data at this higher framerate, we are able to access the fast dynamics occurring at larger wavevectors. We observe that the data more closely follows a $\tau \sim q^{-2}$ relationship, indicating diffusive dynamics.
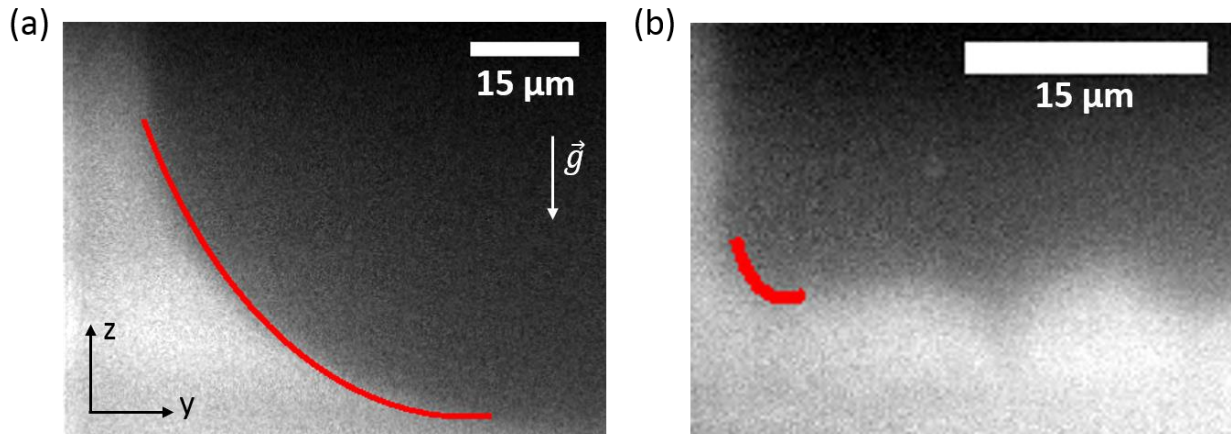
**Figure S4**. Here we show images of the meniscus between the colloid-rich liquid phase and the colloid-poor gas phase. On the left-hand side of both images is a vertical wall that has a wetting layer of the liquid phase. We acquired these images on a custom-built light-sheet microscope where the sample is placed in a 1/32" inner diameter tube and illuminated with a 488-nm laser shaped into a sheet with a thickness in the direction perpendicular to the images shown of ~5 microns. The tube containing the sample is oriented with its long axis along the direction of gravity. We wait >24 hours for the sample to separate into two layers and then image the interface next to the vertical wall of the tube. Following the method described in Aarts[1], we fit the shape of the interface to the equation: $y/L_c = \text{arccosh}(2L_c/z) - \text{arccosh}(2L_c/h) + \left(4 - h^2/L_c^2\right)^{0.5} - \left(4 - z^2/L_c^2\right)^{0.5}$ where $L_c$ is the capillary length, $y$ is the position along the horizontal direction of the interface from the vertical wall, $h$ is the height of the interface at $y=0$, and $z$ is the height above the flat interface. (a) This sample had a composition of 0.043% (w/v) xanthan and a PNIPAM volume fraction of 0.13. We estimate $L_c$ for this sample to be 40 μm. (b) This sample had a composition of 0.024% (w/v) xanthan and a PNIPAM volume fraction of 0.07. We estimate $L_c$ for this sample to be 7 μm.

**Supplementary Video 1**. This video shows 200 frames from a time series of SDIs taken at 0.34 Hz at the interface of a PNIPAM-xanthan sample (PNIPAM volume fraction 0.1; xanthan concentration 0.033% w/v). The video is played back at a rate of 25 fps.

Below, we include the script we run in Micro-Manager which enables us to automatically save the standard deviation of groups of a set number of images.

```
/*
 * Micromanager script used to acquire data
 */

import ij.*;
import ij.gui.*;
import org.micromanager.api.AcquisitionOptions;
import java.lang.System;
import ij.process.*;
import ij.ImagePlus;
import ij.plugin.*;
import java.lang.Math;
import java.awt.image.*;
import ij.measure.*;
import ij.text.*;
import ij.plugin.filter.*;
import org.micromanager.imagedisplay.VirtualAcquisitionDisplay;


// file locations
acqName = gui.getUniqueAcquisitionName("data_50frames");
rootDirName = "C:/Data/";

// Specify number of images to use in calculation of the standard deviation (SD)
nrFrames = 50;  ##here, taking 50 frames and then getting SD of that group
nrChannels = nrSlices = nrPositions = 1;

width = (int) mmc.getImageWidth();  ##width of the image in pixel
height = (int) mmc.getImageHeight();  ## height of the image in pixel
bytesPerPixel = (int) mmc.getBytesPerPixel(); ##bytes per pixel
bitDepth = (int) mmc.getImageBitDepth();  ##bit depth of each image

// Loop over the total number of SD we want to acquire (here, 1000)
for (int i=0; i<1000; i++) {

    //show the images acquired but do not save each individual frame
    gui.openAcquisition(acqName+i, rootDirName, nrFrames, nrChannels, nrSlices, nrPositions,
        /* show */ true,
        /* save */ false);

    gui.initializeAcquisition(acqName+i, width, height, bytesPerPixel, bitDepth);
    mmc.startSequenceAcquisition(nrFrames, 0, true);

    frame = 0;
    exposureMs = mmc.getExposure();    ##exposure time in ms

    while (mmc.getRemainingImageCount() > 0 || mmc.isSequenceRunning(mmc.getCameraDevice())) {
        if (mmc.getRemainingImageCount() > 0) {
            img = mmc.popNextTaggedImage();
            gui.addImageToAcquisition(acqName+i, frame, 0, 0, 0, img);
            frame++;
            }
        else {
            mmc.sleep(Math.min(0.5 * exposureMs, 20));
            }
        }

    mmc.stopSequenceAcquisition();

    //Calculate the standard deviation per pixel for the group of nrFrames frames acquired
    ij.IJ.run("Z Project...", "projection=[Standard Deviation]");

    //Save the result
    IJ.run("Save", "save=C:\\Data\\"+acqName+i+".tif");

    //Close windows
    ij.IJ.run("Close");
    activeWindow = WindowManager.getCurrentWindow();
    VirtualAcquisitionDisplay.getDisplay(activeWindow.getImagePlus()).promptToSave(false);
    ij.IJ.run("Close");

    //Pause until it is time to take next group of images and find the SD of those (here, 1.5 seconds)
    gui.sleep(1500);

}
```

# References

1.  D. G. A. L. Aarts, *J. Phys. Chem. B*, 2005, **109**, 7407-7411.