Dynamic force microscopy in superfluid helium

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Piezoelectric quartz tuning forks have been used for topographic dynamic force imaging in superfluid helium and in high magnetic fields. This has been achieved by immobilizing one tine of the tuning fork to stabilize its behavior in superfluid. Images acquired at room temperature and at 50 K are also presented. Frequency–distance curves are shown to be markedly different in superfluid than in air due to a long-range fork–sample interaction in liquid. Evidence is presented that this is due to a change in the hydrodynamic effective mass of the fork as the gap between the fork and sample is reduced. In addition, Q-control has been implemented and used to both increase and decrease the quality factors of tuning forks in both vacuum and superfluid helium. © 2002 American Institute of Physics. [DOI: 10.1063/1.1496503]

Quartz tuning forks were originally introduced into the field of scanning probe microscopy (SPM) by Günther et al. and later by Karrai and Grober. Recently, Giessibl has employed them for atomic resolution atomic force microscopy (AFM) imaging and King and Nunes have used them to image biomolecules. Tuning forks have previously been used as sensors at low temperatures and in high magnetic fields by Rychen et al. The forks are inexpensive, self-sensing force sensors with high quality factors Q. Their self-sensing capability makes them particularly convenient for low temperature work as the need for more complicated optics inside the cryostat is removed. Thus any problems associated with misalignment of components on cooldown are eliminated. This also makes them especially suited to the study of surfaces that are photosensitive at low temperatures, such as semiconductors. Another advantage for operation at liquid helium temperatures is that tuning forks dissipate very little power (\(\sim pW\)) and therefore do not significantly heat the system under study.

Authors have previously reported difficulty in obtaining images with tuning forks in liquid helium due to fluctuations of the resonant frequency \(f_0\). We have solved this problem by immobilizing one tine of the fork (first proposed by Giessibl for operation at room temperature). Tuning forks are vibrationally symmetric oscillators in which the dynamic forces on the base cancel due to the opposing motions of the tines. However, when an SPM tip is mounted on the end of one tine the symmetry is broken and the forces on the base no longer cancel. Tip–sample interaction forces break the symmetry further. This results in unstable and unpredictable behavior of \(f_0\) and Q. The situation is worsened in liquid due to increased acoustic coupling between the tines, with one tine absorbing acoustic energy radiated by the other. These problems can be avoided by gluing down one tine to a hard, rigid base. The result is effectively a high force constant piezoelectric cantilever. We use nonconducting epoxy resin with fillers to glue the tine onto a metal base.

The tuning fork is excited with an ac voltage at the frequency of its mechanical resonance. The displacement of the piezoelectric crystal induces charge on the electrodes of the tuning fork, and the resulting current is proportional to the velocity of the tine. The current is converted to a voltage by a transconductance amplifier (10^6 V/A) and the phase and amplitude of the voltage are measured with a lock-in amplifier. Excitation amplitudes are typically of the order of 10 mV peak-to-peak. In superfluid helium this results in an rms current of \(\sim 2\) nA on resonance. The mechanical oscillation amplitude can be estimated to within a factor of two from an expression derived in Ref. 3, which gives a theoretical sensitivity of 1.4 nA/mm at 32 kHz for the forks used in this work. Images have also been acquired at drive amplitudes as low as 1 mV peak-to-peak. One advantage of measuring the velocity of the tine is that changes in the damping of the cantilever do not affect the resonant frequency. The resonant frequency is sensitive only to changes in the effective force constant and mass of the sensor.

In the presence of a tip–sample interaction force gradient \(\partial F/\partial z\), \(f_0\) is shifted from \((1/2\pi)(k/m^*)^{1/2}\) to \((1/2\pi)\sqrt{[(k-\partial F/\partial z)/m^*]^{1/2}}\), where \(k\) and \(m^*\) are the effective force constant and mass of the tuning fork tine, respectively. Following other authors in the field, we have implemented a phase-locked loop (PLL) which actively tracks the resonant frequency of the fork so that the fork is always oscillating at its resonant frequency. The PLL can be operated with a sub-ms time constant and can track \(f_0\) to a resolution of better than 10 mHz. Such high frequency resolution is necessary as the sensitivity of a tuning fork to forces and force gradients is reduced by its high force constant. The PLL output \(f_0\) is used by the microscope control electronics to adjust the sample height and thereby keep \(f_0\), and hence the force gradient, constant. Low temperature imaging was performed in a variable temperature insert with a base temperature of 1.2 K inside a vapor-shielded dewar. The main microscope body is a commercial instrument, modified to incorporate tuning fork sensors.

The fork tines used in this work are 3.1 mm long, 0.24 mm wide, and 0.38 mm thick. Their resonant frequency \(f_0\) is nominally 32 768 Hz. The effective force constant \(k\), is \(\sim 8.7\) kN/m. The quality factor Q in air is typically 10 000,
dropping to \(~\sim 2000\) when one tine is immobilized. Standard AFM silicon cantilevers glued to the end of one tine are used as probes. Careful mounting of the tip ensures that the quality factor is not degraded further.

The resonant frequency drops by several hundred hertz on mounting the tip and drops by a similar amount on immersion in superfluid helium. Stable operation was not obtained in liquid helium at temperatures above the superfluid transition due to the boiling of the liquid. Chu\(^{10}\) showed that the frequency of a cantilever beam immersed in an inviscid fluid, \(f_0\), is given by \(f_0 = f_{0e}(1 + \pi n w a/4 \rho t)^{-1/2}\) where \(f_{0e}\) is the resonant frequency in vacuum, \(\rho\) the density of the fluid and \(n\) the density, thickness, and width of the cantilever, respectively. Experimentally we found \(f_{0e} = 32.245\) Hz and \(f_0 = 31.551\) Hz, i.e., a drop in \(f_0\) of 2.15\% on immersion in liquid helium. Taking \(\rho = 145\) kg/m\(^3\) for the density of liquid helium and \(\rho_0 = 2650\) kg/m\(^3\) for quartz, the expression derived by Chu predicts a drop in \(f_0\) of 1.83\%, which is in close agreement with our experimental result.

The acquisition of frequency–distance curves elucidates how the resonant frequency behaves in close proximity to the sample. Such a curve acquired in air is shown in the inset of Fig. 1. Usually when imaging, the frequency setpoint is at how the resonant frequency behaves in close proximity to the wall, its hydrodynamic effective mass \(m'\) increases to \(m' = 1 + 3 r^3/8(r + x)^5\), where \(r\) is the sphere radius and \(x\) is the distance from the wall to the surface of the sphere. If we make the assumption that an expression of similar form is valid for the case of a rectangular beam approaching a surface, we can write the following expression for the resonant frequency of the beam near a surface in an inviscid fluid:

\[
f_0 = \frac{k}{2\pi} \left( \frac{m' + m^* + m'^*}{c^2(a^2/(a + z)^2)} \right)^{1/2},
\]

where \(k\) and \(m^*\) are the force constant and effective mass of the beam, respectively, \(a\) is now a characteristic length, related to the fork dimensions, \(z\) is the tip–sample distance, and \(c\) and \(b\) are constants. The shift of the resonant frequency \(\Delta f\), compared to the frequency of a beam an infinite distance from the surface \(f_{\infty}\), is given by

\[
\Delta f = -\frac{c}{2} f_{\infty} \left( m' + m^* + m'^* \right) \left( a^2/(a + z)^2 \right)^{1/2}.
\]

In fitting the data in Fig. 1 we have assumed this functional form, taking \(a\), \(b\), and \(c\) to be free parameters. \(m^*\) is calculated from the dimensions and density of the tine, and \(m'\) is determined from the change in resonant frequency of the fork on immersion in superfluid. The curve through the data points in Fig. 1 is the best fit to the data, with the parameter values \(a = 130 \pm 60\) \(\mu\)m, \(b = 2.0 \pm 0.8\), and \(c = 0.112 \pm 0.006\). These values are in surprisingly close agreement with those predicted for a sphere. For example, \(a\) is found to be approximately equal to half the width of the tuning fork tine, whereas in the case of a sphere it is the radius. The prefactor \(c\) is of the same order of magnitude as that found for a sphere. \(b\) is slightly lower and this may be due to the different geometries of the two cases. It is believed that the success in fitting the data to this model is strong evidence that the long-range fork–sample interaction is due to the increasing hydrodynamic mass of the cantilever as the gap between the two bodies is narrowed. These effects may become significant for other fluids and cantilevers where \(Q\)-control is used to reduce the width of the resonance of the cantilever. The results reported here will allow researchers to estimate the size of the effect on experiments in other situations and to separate the effects of viscosity from those of the hydrodynamic effective mass. The long-range fork–sample interaction obscures the shorter-range tip–sample interaction and could be avoided by attaching a longer tip (e.g., an etched tungsten wire several hundred microns in length) to the end of the fork. Imaging in superfluid helium is normally performed on the negative slope portion of the frequency distance curve, at a frequency shift of \(~\sim 10\) to 20 Hz above that of the minimum (see Fig. 1).

Figure 2(a) shows a \(6 \mu\)m \(\times\) \(6 \mu\)m image of a silicon calibration grating acquired at room temperature in air. The grating pitch is 3 \(\mu\)m and the step height is 20 nm. Fig. 2(b) shows a similar image acquired at 50 K in vacuum. The \(Q\) of the fork in vacuum is \(~\sim 0\) 000 and such high sensitivity makes microscope control more difficult, as shown by the poorer quality of this image. Figure 2(c) shows a 500 nm \(\times\) 500 nm image of a gold thin film acquired in superfluid helium at 1.7 K and in a magnetic field of 10 T. Imaging could also be performed while the field was being swept...
from 0 to 10 T, with negligible shift in the relative lateral

tip–sample position. The most important point is that stable
topographic imaging utilizing a piezoelectric quartz tuning
fork in superfluid helium has been achieved. This success is
due to the fact that one tine has been immobilized, as
discussed earlier.

We also report the use of $Q$-control to tune the quality
factor of the tuning forks. Tamayo et al.\cite{15} have used
$Q$-control while imaging soft biological samples in liquid to
increase cantilever $Q$ values so that a lower drive force can
be used, resulting in less sample damage. Sulchek et al.\cite{16}
have used $Q$-control to enable faster scanning by reducing $Q$
values, hence reducing the mechanical settling time of the
cantilever. A schematic of the circuit can be seen in Fig. 3(a).

As can be seen, the tuning fork response is phase shifted and
amplified before being added to the drive. The effective
damping is altered by feeding back some of the response, with
the phase of this feedback signal, relative to the fork
velocity, determining whether the damping is effectively in-
duced or decreased. The degree of alteration of the damp-
ing is altered by feeding back some of the response,
amplified before being added to the drive. The effective

In summary, we have demonstrated topographic imaging
in superfluid helium and in high magnetic fields using dy-
namic force microscopy with a piezoelectric quartz tuning
fork as sensor. This was made possible by immobilizing one
tine of the tuning fork, so that $f_0$ and $Q$ are better behaved and the inter-tine acoustic coupling is reduced. The observed
long-range fork–sample interaction in superfluid can be ex-
plained in terms of a change in the hydrodynamic effective
mass of the tine as the fork–sample gap is reduced. It has
been shown that $Q$ values of tuning forks can be altered
(increased or decreased) significantly using the technique of
$Q$-control.

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\begin{figure}
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\includegraphics[width=\textwidth]{figure2}
\caption{Topographic images of (a) silicon grating acquired in air at room
temperature, (b) silicon grating acquired in vacuum at 50 K, (c) gold thin
film acquired in superfluid helium at 1.7 K and in a magnetic field of 10 T.
(d) is a cross section taken along dotted line in (c).}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure3}
\caption{(a) Schematic of $Q$-control feedback circuit and (b) the result of
using $Q$-control in superfluid helium to increase and decrease the $Q$
of a tuning fork with one tine immobilized.}
\end{figure}

\begin{thebibliography}{16}
\bibitem{3} F. J. Giessibl, Appl. Phys. Lett. 76, 1470 (2000).
\bibitem{5} J. Rychen, T. Ihn, P. Studerus, A. Herrmann, and K. Ensslin, Rev. Sci.
Instrum. 70, 2765 (1999).
\bibitem{6} F. J. Giessibl, Appl. Phys. Lett. 73, 3956 (1998).
\bibitem{7} JB Weld, JB Weld Company Ltd., UK (www.jbweld.co.uk).
\bibitem{9} CryoXSM, Oxford Instruments plc, UK (www.oxinst.com).
(Southwest Research Institute, San Antonio, TX, 1963).
\bibitem{11} S. J. O’Shea and M. E. Welland, Langmuir 14, 4186 (1998).
\bibitem{13} J. Tamayo, A. D. L. Humphris, R. J. Owen, and M. J. Miles, Biophys.
\bibitem{14} L. M. Milne-Thomson, Theoretical Hydrodynamics, 5th edition (Mac-
\bibitem{15} J. Tamayo, A. D. L. Humphris, and M. J. Miles, Appl. Phys. Lett. 77, 582
(2000).
\bibitem{16} T. Sulchek, R. Hsieh, J. D. Adams, G. G. Yaralioglu, S. C. Minne, C. F.
Quate, J. P. Cleveland, A. Atalar, and D. M. Adderton, Appl. Phys. Lett.
76, 1473 (2000).
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