Fabrication of single-mode chalcogenide fiber probes for scanning near-field infrared optical microscopy

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Abstract. We fabricate scanning near-field optical microscope (IR-SNOM) probe tips made from singlemode chalcogenide fiber and test them using a standard SNOM setup and free-electron laser. SEM micrographs, showing tips with submicrometer physical dimensions, demonstrate the feasibility of the thermal micropipette puller process used to create the tips. Topographical data obtained using a shear-force near-field microscope exhibit spatial resolution in the range of 80 to 100 nm. Optical data in the IR (near 3.5 μm), using the probe tips in collection mode, indicate an optical spatial resolution of approximately $\lambda/15$.
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Scanning near-field optical microscopy (SNOM) has been shown to have great utility for applications including imaging and analysis of semiconductor devices,[1–3] microstructural spectroscopy of organic films,[4,5] and biological research.[6,7] Yet little work has been done in the mid- to long-IR (from 2 to 11 μm) (although some work has been done at 10.6 mm using CO_2 lasers as in Lahrech et al.[8]), largely because there are three diverse skill areas that must coincide: expertise in near-field microscopy, expertise in and possession of high-power mid-IR sources such as free-electron lasers (FELs), and expertise in IR fibers as well as the ability to make high quality fiber and taper it into probes. As yet, only multimode fiber has been used[9] for IR-SNOM,[9] with spatial resolution approaching 1 μm or about $\lambda/6$. In this paper, we report on the fabrication of SNOM probes from singlemode chalcogenide fiber, and the performance of those probes in measurements using the Vanderbilt FEL, where optical images that suggest a resolution approaching $\lambda/35$ were obtained.

These IR SNOM (IR-SNOM) probes were made from singlemode arsenic sulfide fiber (with an outside diameter of 80 to 140 μm and a core of approximately 10 μm), fabricated and tested at the Naval Research Laboratory as described elsewhere.[10] The tips were made using a standard micropipette puller (Sutter P-2000), with either a heated filament or an external visible laser used to soften the fiber. The laser source supplied with the pipette puller would not work in this case, since the chalcogenide fiber is transparent to CO_2 radiation. In addition, the 160 g static tension of the micropipette puller was often enough to break the thin-diameter chalcogenide fiber, so a counterbalance system had to be devised to relieve some of the static load. As described here, the detection of fiber velocity plays a crucial role in making microtips in this way, so care was taken to use the minimum amount of weight for the counterbalancing.

Variations on standard silica fiber tip geometry from the parameters of velocity, delay, and pull strength available through this pipette puller's interface have been described at length in other papers,[11,12] so we simply state that we used a standard matrix-experimentation approach when searching this parameter space for suitable chalcogenide settings. Note that our parameter set also included variability in laser/heater intensity and counterweighting.

The tip fabrication techniques studied can be grouped into two categories: laser-heated and furnace-heated. The laser heating experiment was undertaken first, using the 514 nm line of an argon laser focused by a cylindrical lens onto the fiber. Though many chalcogenide glasses exhibit a strong thermal runaway effect with laser heating, due to a large positive increase in the absorption coefficient with temperature,[13] it was hoped that the softening and subsequent thinning of the fiber would provide a good limit to this process. However, we observed a sharp threshold in laser power for tip formation: below a certain level, the fiber would not soften at all; above that level, the softening and breakage were almost instantaneous. As shown in Fig. 1, the tips produced by this method, although often of very

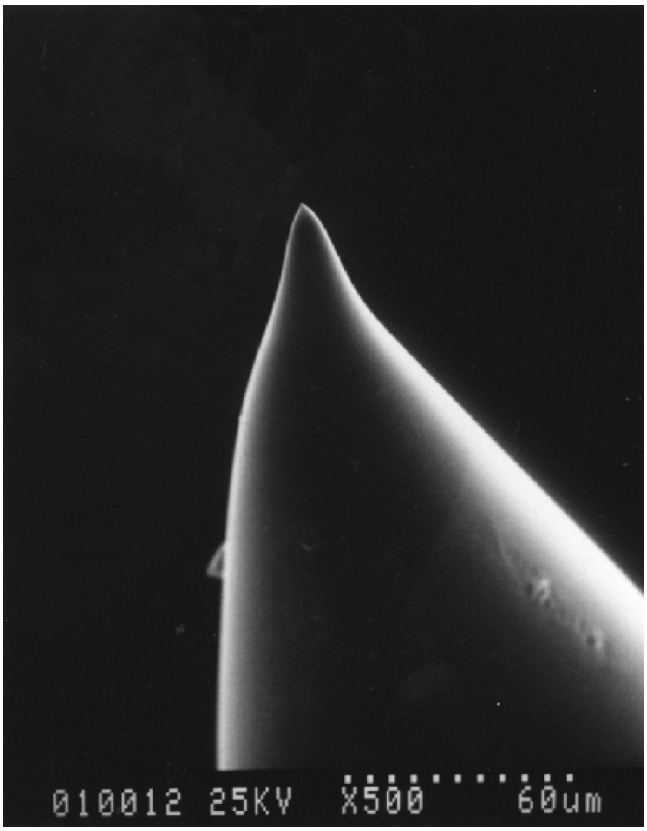


Fig. 1 Laser-heated singlemode chalcogenide fiber tip. The scarring seen at the right is most likely due to excessive lateral recoil (due to the plastic deformation described in the text) and subsequent glancing impact with the furnace.

small radius, were very asymmetric, exhibiting much greater melting effects on the side of the fiber exposed to the laser. Simple retroreflection and retrofocusing of the laser beam did not alleviate this problem. Unfortunately, it does not appear possible to control the thermal runaway without some means of regulating the optical dose put into the fiber.

To fabricate the tips using a thermal source, small wound heaters of approximately 6 mm inside diameter were constructed from standard nichrome wire; the optimum length of the hot zone was determined to be about 3 mm. The heating system was similar to one we described previously for making chalcogenide fiber couplers.[14] Counterweighting was adjusted to inhibit static breakage of the fiber, which was completely stripped of its coating and cleaned with solvents.

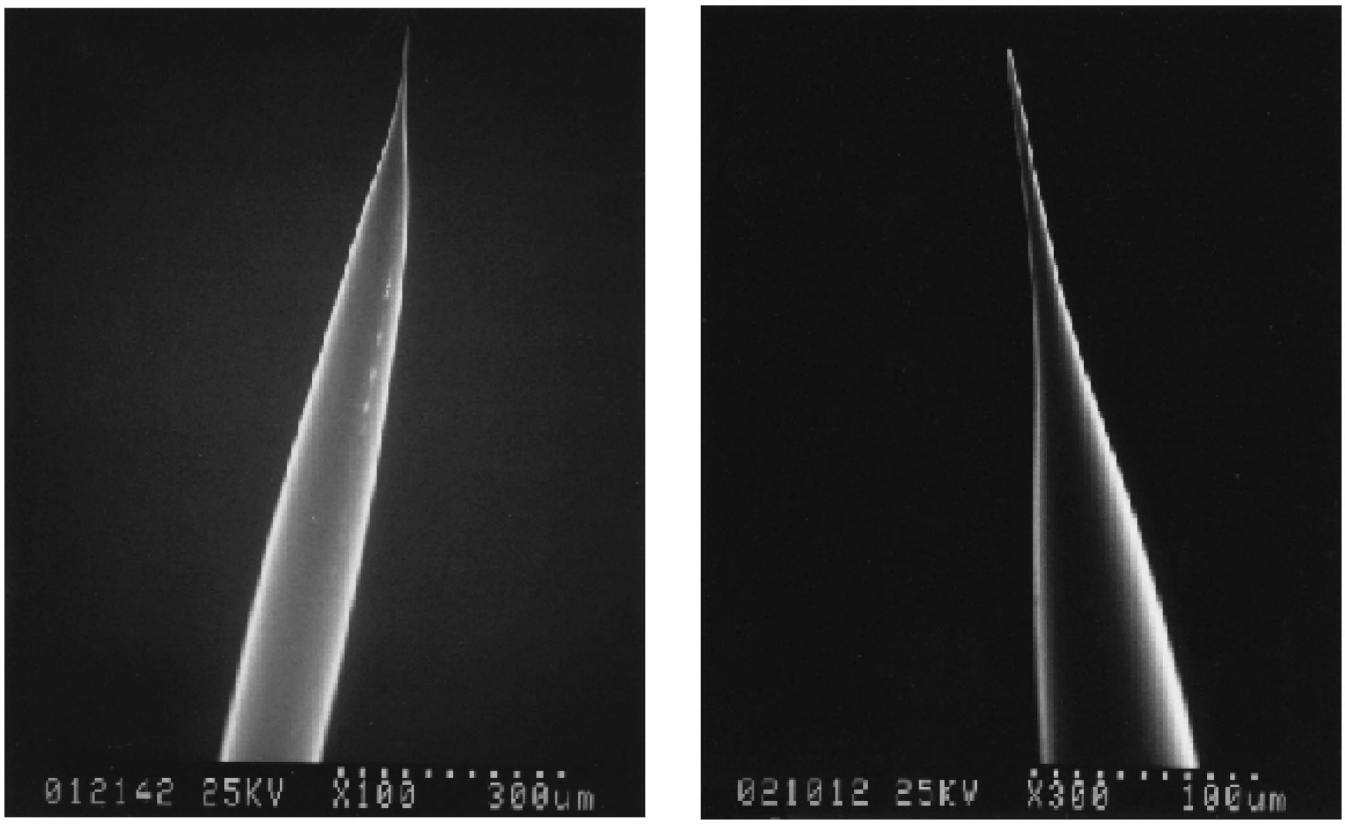
The most significant influence on tip formation was found to be the heater current, controlling both the heating rate and the ultimate furnace temperature. This makes intuitive sense if one considers how the fiber softens and flows to allow good tips to form under a given load and is determined largely by how the viscosity of the glass changes in time.[15] In chalcogenides, the slope of the viscosity versus temperature curve is steeper than in silica,[16] so the range of temperature control is correspondingly narrower. At low currents, sufficient just to soften the fiber and enable viscous flow, the resulting tapers were rounded at very large diameters and had no useable tips. This indicates that the fiber did not soften over a large enough area to promote even flow and thus a smooth taper and a good tip. At high currents, where the onset of flow occurred very quickly, the tapers were often narrow and of very small tip diameter, but the viscosity of the fiber was low enough that the tapers deformed or curled more often than not. This deformation was exacerbated by the viscoelastic deformation that some of the fibers had undergone from being stored on a spool for more than a year, which led to exaggerated lateral recoil when the two parts of the fiber separated [as exemplified in Fig. 2(a)].

Using a suitable heater setting, the next biggest influence on tip formation was the velocity parameter of the puller, essentially the viscosity trip-point where the harder solenoid pull would be initiated. As has been found with silica fiber,[1] lower velocity settings yielded better results, down to a minimum where the puller could not measure the velocity accurately. We also made several pulls with a maximal velocity setting and no solenoid pull to observe the result of a pure ''gravity pull,'' but these tips tended to be overly long and deformed. With velocity and heat settings as described, the strength of the solenoid pull could be adjusted over a relatively large range to give good results. The delay parameter had little or no observed effect.

Smooth tapers with very small tips, typified by the one shown in Fig. 2(b), were obtained in this manner. No statistically significant correlation was obtained between the tip diameters and the pulling parameters, but the qualitative results, such as those shown in Fig. 3, were statistically reproducible. Several microtips were made with diameters estimated at 100 to 200 nm.

However, the apparent physical diameter of the tip may not be an accurate reflection of what spatial resolution it will give, and in any event, actual performance numbers are more relevant. As discussed often in the SNOM literature, the resolution of the probe can be highly dependent on the topography of the specimen.[6] To gauge the performance of our chalcogenide SNOM probes, we tested them in a microscope setup constructed at the Istituto Struttura Materia[17,18] (ISM-CNR) in Rome for use with the Vanderbilt FEL.

The specimens examined in this study were polycrystalline diamond films prepared by plasma chemical vapor-phase deposition.[19] The SNOM was first calibrated using standard silica fiber tips, then several topographical (shear-force) images were taken with the chalcogenide tips to verify that the data from the new tips correlated with the old. As shown in Fig. 4, the chalcogenide tips were able to resolve even small features on the individual grains quite well. From these small features, we estimated the best lateral resolution of the probe (from the FWHM of diagonal cross sections) to be between 50 and 80 nm. Since the surface was quite rough (at least on a nanoscopic scale), these images indicate that the probe aperture was somewhat smaller than our physical estimates. Also, since irregularities in the probe are expected to be a large detriment to lateral resolution on rough specimens,[6,20] these topographical images indicate that the probe tip itself was smoothly rounded.



(a) (b)

Fig. 2 Thermally produced microtips in singlemode chalcogenide fiber. The tip at the left shows a slight bending due to lateral recoil, while the tip on the right is straight and symmetric. The tip diameters are approximately 500 nm (left) and 150 nm (right).

Optical images were taken simultaneously with the topographical scans, selecting the C-H stretch mode at 3.5 μm as the target image wavelength, with the FEL illuminating the specimen over a broad area and the SNOM probe collecting the reflected light. Figure 5 shows a pair of images obtained in this manner, where the correlation between optical data and certain features in the topographical image can be seen clearly. The topographical structure was reproduced with the FEL tuned outside the C-H absorption band (to 3.2 or 3.7 μm), but the optical image became featureless, indicating that the dark regions in the optical image of Fig. 5 correspond to regions where the absorption was stronger. Though the relative intensity noise in the FEL (which could not be normalized out in the apparatus used for this paper) is evident in Fig. 5, several small features and edges are still resolved. Though the small features were reproducible and would suggest an optical lateral resolution to be between 100 and 150 nm (about $\lambda/25$ to $\lambda/35$), a more conservative estimate of the resolution would be $\lambda/15$.

These results demonstrate clearly that singlemode chalcogenide fiber can yield excellent results in this novel FEL-SNOM combination. As this was the first attempt at such an experiment and many of the systems and indeed the probes themselves had not been fully optimized, we expect that these performance numbers will be surpassed in the near future. Applications for IR-SNOM, especially at wavelengths longer than about 2 μm where silica fiber does not transmit, range from studies of buried semiconductor interfaces and implants to imaging of subcellular features in animal tissues. As interest grows in obtaining high resolution images of inorganic and biological specimens with distinguishing spectral features in the mid- to long-IR region, these probes could prove an indispensable research tool.

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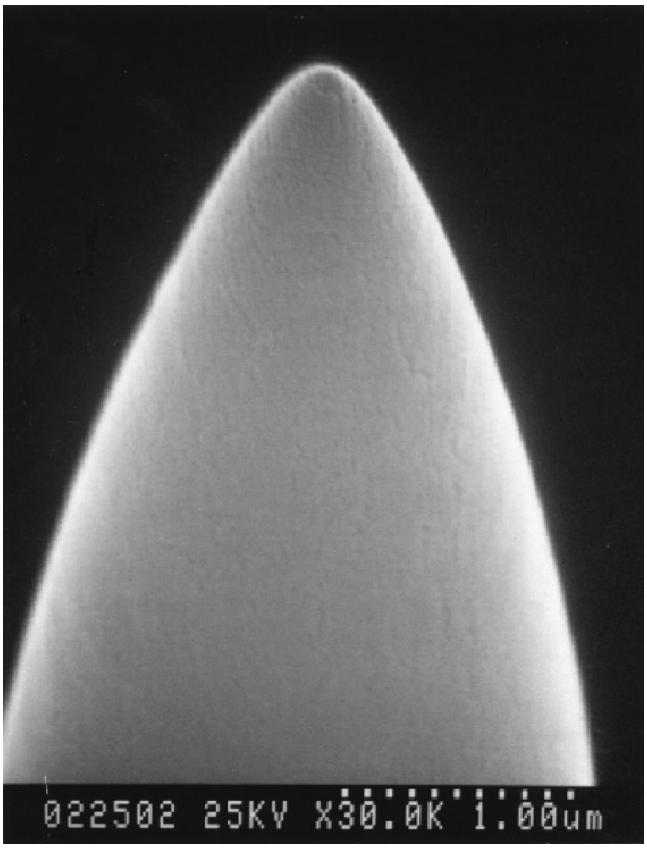

Fig. 3 High-magnification image of chalcogenide fiber SNOM tip. The variegation on the surface is due to the metal coating used for scanning electron microscopy (SEM) preparation.

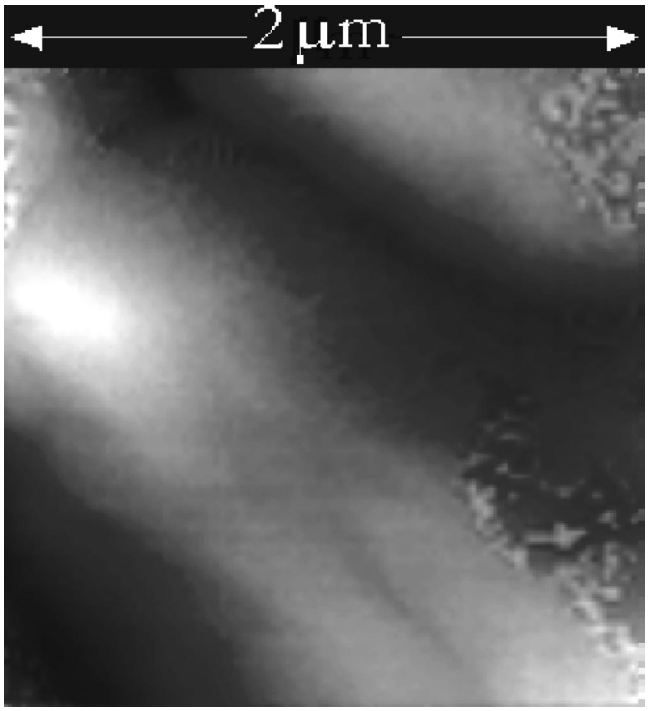

Fig. 4 Topographical (shear-force) image of diamond grains obtained with chalcogenide SNOM tip. The mottled features at the lower and upper right and upper left are most likely impurities or imperfections in the grains. Each scan step was 10 nm; the entire image is 200×200 points.

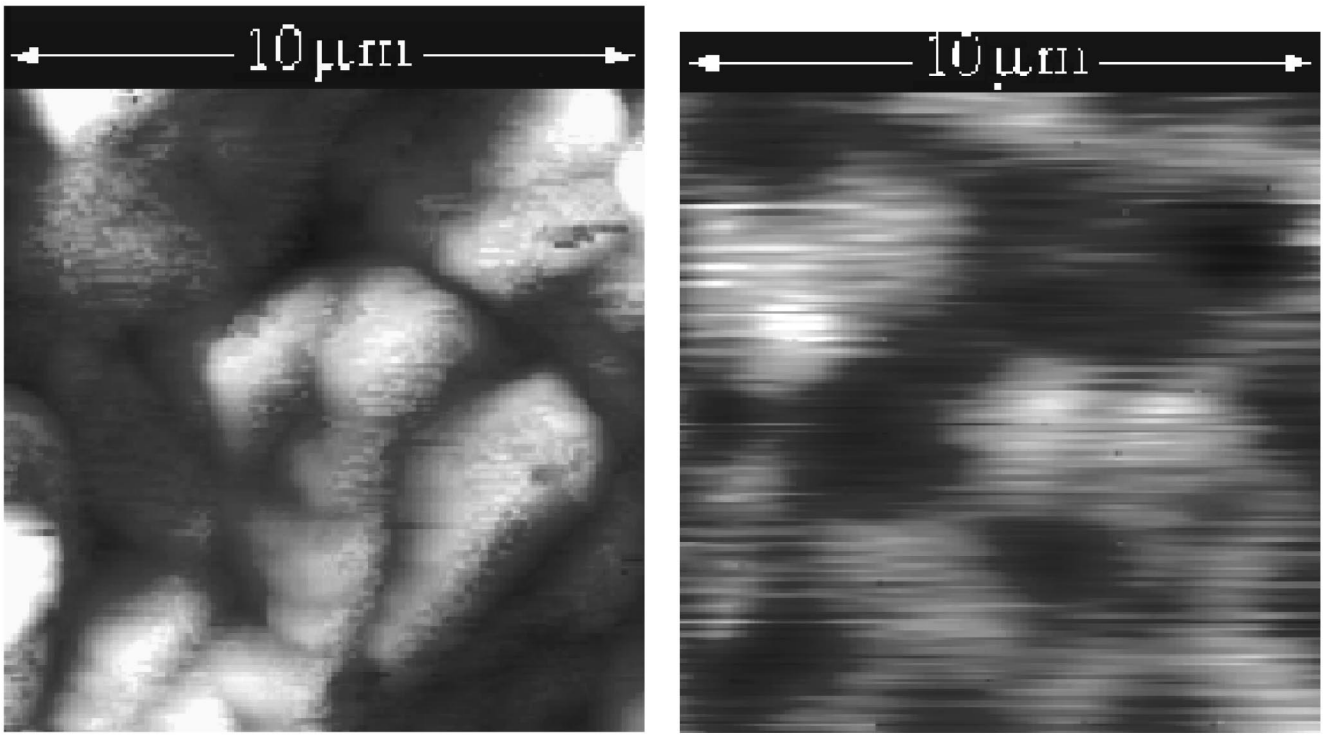

Fig. 5 Comparison of simultaneous topographical (left) and optical (right) images taken with the IR-SNOM. The dark areas in the optical image represent areas where the C-H vibrational absorption was strongest. These optical features were evident only when the FEL was tuned into the C-H absorption band around 3.5 μm.

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