PROCEEDINGS OF THE TWENTY-EIGTH CONFERENCE OF THE NORTH AMERICAN THERMAL ANALYSIS SOCIETY, OCTOBER 4-6, 2000, SAVANNAH, GEORGIA

THERMAL CONDUCTIVITY IMAGING USING THE ATOMIC FORCE MICROSCOPE

Duncan M. Price & Mike Reading
IPTME, Loughborough University, Loughborough, Leics. LE11 3TU, UK
d.m.price@lboro.ac.uk

Azzedine Hammiche & Hubert M. Pollock
School of Physics & Chemistry, Lancaster University, Lancaster LA1 4YB UK

ABSTRACT

Scanning thermal microscopy employs a miniature temperature sensor such as a thermocouple or resistance thermometer in place of the conventional inert sharp tip used in atomic force microscopy. This can be rastered over the surface of a specimen to map its temperature distribution in a purely passive mode in order to detect hot spots in semiconductor devices. By controlling the tip's temperature (either by illuminating the tip with a laser or by Joule heating arising from passing a current through the tip) the heat flow from the tip to the surface can be used to obtain images whose contrast depends upon the specimen's thermal transport properties (thermal conductivity and thermal diffusivity). AC heating of the tip affords a means to perform 3 dimensional tomographic imaging of the sample due to the thermal diffusion length of the evanescent temperature wave being dependent on its frequency. Applications and limitations of the technique are discussed along with progress towards quantitative measurement of thermal properties using this approach.

INTRODUCTION

The inventions of the scanning tunneling microscope (STM) *(1)* and the atomic force microscope (AFM) *(2)* have allowed sub-micrometer and, at times, atomic scale spatially resolved imaging of surfaces. Spatially-resolved temperature measurements using optical systems are diffraction limited by the wavelength of the radiation involved which is about 5-10 µm for infrared thermography and about 0.5 µm for visible light. The spatial resolution of near-field techniques (such as AFM) is only limited by the active area of the sensor (which in the case of STM may only be a few atoms at the end of a metal wire).

The first experiments in scanning thermal microscopy (SThM) were carried out by Williams and Wickramasinghe who employed a heated thin-film thermocouple fabricated from a conventional STM tip *(3)*. As the tip approached a surface it was cooled due to tip-substrate heat transfer. By using the temperature sensed by the thermocouple as a feedback to maintain a constant tip-substrate gap, this scanning thermal profiler could overcome the limitations of STM and be used to image electrically insulating surfaces with a lateral resolution of 100 nm. Since the feedback signal was based on maintaining a constant probe temperature, it could not be used to obtain true thermal images of surfaces –

instead it effectively measured the topography of the specimen from the increase in heat flux as the tip neared the surface.

In an attempt to overcome the limitations of this method of SThM, Majumdar described the use of an AFM cantilever fashioned from a pair of dissimilar metal wires (Chromel and Alumel) which met to form a thermocouple junction at the tip *(4)*. In this way, the conventional AFM force feedback mechanism could be used to measure surface topography whilst at the same time mapping the temperature distribution of energized electronic devices with sub-micrometer resolution. Since this demonstration, a number of different probe designs have been developed and progress has been made towards the measurement of absolute thermal conductivities and 3-dimensional tomographic imaging. It is the intention of this paper to reflect on these developments, whereas more general coverage of SThM can be found in the reviews by Gmelin *(5)* and Majumdar *(6)*.

THERMAL ELEMENT DESIGN

Three techniques have been used to combine the conventional AFM cantilever with a means of localized thermometry:

1. Thermocouple Cantilevers

The use of SPM sensors with a thermoelectric element at the tip has been described above. In an effort to improve the performance of a bare thermocouple tip, Majumdar *et al.* cemented a diamond shard to the junction so as to give a harder tip with improved spatial resolution and reduced thermal resistance *(7)*. The same group also describe depositing successive layers of different metals so as to make thermocouple pair on top of a standard "A-frame" AFM cantilever *(8)*. Fish *et al.* borrowed from near-field scanning optical microscopy technology to make a thermocouple derived from gold-coated glass micropipettes containing a platinum core *(9)*. Workers at Glasgow University have fabricated thermocouple probes using electron beam lithography and silicon micromachining in order to deposit one or more thermocouple junctions at the AFM tip *(10,11)*. Such work leads to the possibility of building thermopile sensors (perhaps with the incorporation of a heater) analogous to a miniature heat flux calorimeter.

2. Resistance Thermometry

In 1994 Dinwiddie and Pylkki described first combined SThM/AFM probes that employed resistance thermometry to measure thermal properties *(12,13)*. These were fashioned made from Wollaston process wire. This consists of a thin platinum core (ca. 5 μm in diameter) surrounded by a thick silver sheath (ca. 75μm). A loop of wire is formed and the silver is etched away to reveal a small length of platinum which acts as a miniature resistance thermometer. This can be operated in two modes: a) as a passive thermosensing element (by measuring its temperature using a small current) or b) as an active heat flux meter. In the latter

case, a larger current (sufficient to raise the temperature of the probe above that of the surface) is passed through the probe. The power required to maintain a constant temperature gradient between the tip and sample is monitored by means of an electrical bridge circuit. In essence, this is equivalent to a power compensation calorimeter. Mills *et al.* describe similar probes in which the resistance element is deposited across the apex of a silicon nitride pyramid similar to a conventional AFM cantilever *(14)*. Both designs of sensor are commercially available.

In a passive mode, such devices function like thermocouple probes described above. These can be used (for example) to map the temperature distribution in energized electronic devices simultaneously with their topography. If the surface is illuminated with infrared radiation, the photothermal effect arising from the absorption of energy specific to the IR active modes of the specimen may be used to obtain the sample's IR spectrum *(15)*. In the active mode, the heat flow from the tip can be used to detect surface and subsurface defects of different thermal conductivity than the matrix *(14,16)*. Furthermore, by placing the probe at selected locations on the sample, its temperature can be changed in a controlled fashion so that the power required to heat the tip can be used to carry out a form of spatially resolved calorimetry *(17,18)*. In addition, since the probe position is monitored by the AFM stage, localized thermomechanical measurements may be carried out concurrent with the calorimetry *(19)*. Binnig *et al.* have inverted this approach and used the indentations in a polymer substrate produced by localized Joule heating of a resistive probe (or array of probes) as a means of high density data storage *(20)*.

3. Bimetallic Sensors

Nakabeppu *et al.* described the use of composite cantilevers made from tin or gold deposited on conventional silicon nitride AFM probes to detect spatial variations in temperature across an indium-tin-oxide heater *(21)*. Differential thermal expansion of the bimetallic elements causes the beam to bend. This deflection is detected using the AFM. In order to separate thermal deflection of the beam from displacement of the cantilever caused by the sample topography, an intermittent contact mode of operation was employed. Measurements were made under vacuum so as to minimize heat loss. A more practical use of this technology is in the form of miniature chemical and thermal sensors *(22)*. This approach has been used to perform thermal analysis on picoliter volumes of material deposited on the end of a bimetallic cantilever *(23)*. Arrays of such devices have applications as sensitive electronic "noses".

THERMAL CONDUCTIVITY MEASUREMENTS AT THE NANOSCALE

Small-scale measurements would benefit the semiconductor and other industries where thermal transport properties are significantly different to and cannot be inferred from measurements at higher scales. Examples of key areas of modern technology and science which might be expected to benefit include microelectronics, cellular biology, forensics, pharmaceuticals, polymer science

etc. In theory, heated thermal probes are capable of measuring the absolute thermal conductivity of materials by the heat flux between the tip and the surface. In practice, heat losses also occur both within the probe and to the atmosphere. Furthermore, the contact area between the tip and the specimen is usually unknown. Ruiz *et al.* developed a simple method for converting heat flux to thermal conductivity by using hard materials of known thermal conductivity to calibrate the system *(25)*. This procedure was used to determine the thermal conductivity of diamond-like nanocomposites to a precision of ±15%. Gorbunov *et. al.* measured the change in heat flux as the probe approached the sample surface or was ramped in temperature in contact with the specimen so as to derive its thermal conductivity – again by calibration with samples of known response *(26,27)*. Fiege *et. al.* used AC heating of the tip to measure the thermal conductivity of silver and diamond using gold as a single point reference material in order to estimate the contact area of the tip *(28)*. One major problem in this area is the deconvolution of the effects of surface roughness upon the thermal conductivity contrast – sharp changes in the slope of the surface (such as at a ridge or valley on the sample) change the contact area of the tip *(16)*. It is therefore useful to compare the topographic image with the thermal image in order to determine what features in the latter are due to true spatial variations in thermal conductivity.

3-D TOMOGRAPHIC IMAGING

The decay length of thermal waves produced by AC heating of a tip varies as a function of the reciprocal of its frequency. Thus it is possible to detect variations in thermal response at shallower depths by using a high frequency temperature modulation superimposed on the conventional DC heating of the tip *(19)*. Several groups have employed this technique to study the thermal diffusivity variations in materials *(17,28,29)*. Gomès *et al.* have theoretically examined this process *(30)* and there is great potential for the use of multiple frequency modulated-temperature SThM as a means to provide non-destructive three dimensional imaging of optically opaque samples using similar principles to those employed for medical imaging by electrical impedance tomography *(31)*.

CONCLUDING REMARKS

This paper presents a brief overview of the technology of scanning thermal microscopy, its applications, limitations and potential. Details of recent progress in the field of localized physical and chemical characterization using thermal probes can be found elsewhere *(32-34)*. AFM is rapidly gaining acceptance in all areas of materials characterization. The growing commercial availability of SThM instrumentation will broaden its scope of usage and lead to a better understanding of the mechanisms of heat transport from the tip to the surface. This can be expected to make routine measurements of absolute thermal properties possible. The thermodynamic limit of measurement (kT) is about 10^{-21} J at room temperature *(7)*. The spatial resolution of STM is around 10^{-10} m. The maximum temperature resolution of the most sensitive thermal probes (bimetallic cantilevers) is 10^{-5} K with an estimated sensitivity limit of 10^{-12} J and a spatial

resolution of 10 $^{-7}$ m. Improvements in both sensitivity and resolution can be expected and presents intriguing challenges for the future.

REFERENCES

1. G. Binnig and H. Rohrer; *Helv. Phys. Acta.*, **1982** 55 726-735.
2. G. Binnig, C.F. Quate and Ch. Gerber; *Phys. Rev. Lett.***1986** 56(9) 930-933.
3. Williams, C.C. and Wickramasinghe. H.K., *Appl. Phys. Lett.*, **1986** 49(23) 1587-1589.
4. A. Majumdar, J.P. Carrejo and J. Lai; *Appl. Phys. Lett.* **1993** 62(20) 2501-2503.
5. E. Gmelin, R. Fisher and R. Stitzinger; *Thermochim. Acta* **1998** 310 1-17.
6. A. Majumdar; *Ann. Rev. Mat. Sci.* **1999** 29 505-585.
7. A. Majumdar, J. Lai, M. Chandrachood, O. Nakabeppu, Y. Wu and Z. Shi; *Rev. Sci. Instrum.* **1995** 66(6) 3584-3592.
8. K. Lou, Z. Shi, J. Varesi and A. Majumdar; *J. Va. Sci. Technol. B* **1997** 15(2) 349-360.
9. G. Fish, O. Bouevitch, S. Kokotov, K. Lieberman, D. Palanker, I. Turovets and A. Lewis; *Rev. Sci. Instrum.* **1995** 66(5) 3300-3306.
10. G. Mills, H. Zhou, A. Midha, L. Donaldson and J.M.R. Weaver; *Appl. Phys. Lett.* **1998** 72(22) 2900-2902.
11. H. Zhou, G. Mills, B.K. Chong, A. Midha, L. Donaldson and J.M.R. Weaver; *J. Vac. Sci. Technol. A* **1999** 17(4) 2233-2239.
12. R.B. Dinwiddie, R.J. Pylkki and P.E. West; in T.W. Tong (editor), *Thermal Conductivity* 22, Technomics, Lancaster PA, **1994**, pp. 668-677.
13. R.J. Pylkki, P.J. Moyer and P.E. West,; *Jpn. J. Appl. Phys.*1 **1994** 33(6B) 784-3790.
14. G. Mills, J.M.R. Weaver, G. Harris, W. Chen, J. Carrejo, L. Johnson and B. Rogers; *Ultramicroscopy* **1999** 80(1) 7-11.
15. A. Hammiche, H.M. Pollock, M. Reading, M. Claybourn, P.H. Turner and K. Jewkes; *Appl. Spectroscopy* 53 (7) **1999** 810-815.
16. A. Hammiche, H.M. Pollock, M. Song and D.J. Hourston; *Meas.Sci. Technol.* 7 **1996** 142-150.
17. A. Hammiche, H.M. Pollock, M. Reading and M. Song; *J. Vac. Sci. Technol. B* **1996** 14(2) 1486-1491.
18. D.S. Fryer, J.J. dePablo and P.F. Nealey; *Proc. SPIE* **1998** 3333 1031-1039.
19. M. Reading, D.J. Hourston, M. Song, H.M. Pollock and A. Hammiche; *Am. Lab.* **1998** 30(1) 13-17.
20. G. Binnig, M. Despont, U. Drechsler, W. Häberle, M. Lutwyche, P. Vettiger, H.J. Mamin, B.W. Chui and T.W. Kenny; *Appl. Phys. Lett*. **1999** 74(9) 1329-1331.
21. O. Nakabeppu, M. Chandrachood, Y. Wu, J. Lai, J. and A. Majumdar; *Appl. Phys. Lett.* **1995** 66(6) 694-696.
22. J.K. Gimzewski, Ch. Gerber, E. Meyer and R.R. Schlittler; *Chem. Phys. Lett.* **1994** 217(5-6) 589-594.
23. R. Berger, Ch. Gerber, J.K. Gimzewski, E. Meyer and H.J. Güntherodt; *Appl. Phys. Lett.* **1996** 69(1) 40-42.

24. F. Riuz, W.D. Sun, F.H. Pollak and C. Venkatraman,; *Appl. Phys. Lett.* **1998** 73(13) 1802-1804.
25. V.V. Gorbunov, N, Fuchigami, J.L. Hazel and V.V. Tsukruk; *Langmuir* **1999** 15(24) 8340-8343.
26. V.V. Gorbunov, N, Fuchigami and V.V. Tsukruk; *Probe Microscopy* **2000** in press.
27. G.B.M. Fiege, A. Altes, R. Heiderhoff and L.J. Balk; *J. Phys. D: Appl. Phys.* **1999** 32 L13-L17.
28. E. Oesterschultze, M. Stopka, L. Ackerman, W. Sholtz and S. Werner; *J. Vac. Sci. Technol. B.* **1996** 14(2) 832-837.
29. L.J. Balk, M. Maywald and R.J. Pylkki.; *9th Conf. on Microscopy of Semiconducting Materials (Inst. Phys. Conf. Ser. 146)*, Oxford **1995** pp. 655-658.
30. S. Gomès, F. Depasse and P. Grossel; *J. Phys. D.: Appl. Phys.* **1998** 31(19) 2377-2387.
31. P. Metherall, D.C. Barber, R.H. Smallwood and B.H. Brown; *Nature* **1996** 380 509-512.
32. D.M. Price, M. Reading, A. Hammiche and H.M. Pollock; *Int. J. Pharm.* **1999** 192(1) 85-96.
33. A. Hammiche, M. Conroy, G.B. Mills, G. B., D.M. Price, M. Reading and H.M. Pollock; *J. Vac. Sci. Technol.* B **2000** 18(3) 1322-1332.
34. D.M. Price, M. Reading, A. Hammiche and H.M. Pollock; *J. Thermal. Anal. Cal.***2000** 60(3) 723-733