CONF-770230--1

THE FEASIBILITY OF ACHIEVING 15 nm RESOLUTION WITH A
SCANNING MICROPROBE USING A LIQUID GALLIUM ION SOURCE

V. E. Krohn and G. R. Ringo

Prepared for

Workshop on Short Wavelength Microscopy
New York, New York
February 23-25, 1977

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

UFC-ANA-USERDA

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

leg

operated under contract W-31-109-Eng-38 for the
U. S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

The facilities of Argonne National Laboratory are owned by the United States Government. Under the terms of a contract (W-31-109-Eng-38) between the U. S. Energy Research and Development Administration, Argonne Universities Association and The University of Chicago, the University employs the staff and operates the Laboratory in accordance with policies and programs formulated, approved and reviewed by the Association.

MEMBERS OF ARGONNE UNIVERSITIES ASSOCIATION

The University of Arizona	Kansas State University	The Ohio State University
Carnegie-Mellon University	The University of Kansas	Ohio University
Case Western Reserve University	Loyola University	The Pennsylvania State University
The University of Chicago	Marquette University	Purdue University
University of Cincinnati	Michigan State University	Saint Louis University
Illinois Institute of Technology	The University of Michigan	Southern Illinois University
University of Illinois	University of Minnesota	The University of Texas at Austin
Indiana University	University of Missouri	Washington University
Iowa State University	Northwestern University	Wayne State University
The University of Iowa	University of Notre Dame	The University of Wisconsin

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights. Mention of commercial products, their manufacturers, or their suppliers in this publication does not imply or connote approval or disapproval of the product by Argonne National Laboratory or the U. S. Energy Research and Development Administration.

3/9/77

THE FEASIBILITY OF ACHIEVING 15 nm RESOLUTION WITH A
SCANNING MICROPROBE USING A LIQUID GALLIUM ION SOURCE*

V. E. Krohn and G. R. Ringo

Argonne National Laboratory, Argonne, Illinois 60439

By acceptance of this article, the publisher or recipient acknowledges the U. S. Government's right to use in a nonexclusive, royalty-free license in and to any copyright material containing the article.

The scanning ion microprobe with secondary-ion mass analyzer^{1, 2} is capable of locating specific stable isotopes in experimental samples. In this role, it competes not with the electron microscope, but with radioautography, a technique which does make use of the electron microscope but which is mainly limited by the shortcomings of radioautography itself. The potential advantages that the ion microprobe has over radioautography include the following:

1) The resolution limit of the ion microprobe is set by the volume from which secondary ions are likely to originate in the neighborhood of a particular incident ion. This volume almost certainly has a diameter which is less than 10 nm and a depth of less than 2 nm.³ On the other hand, the resolution limit of radioautography is ordinarily determined by a beta-ray range of approximately 100 nm or more.⁴ The ion microprobe has a clear advantage here if its resolution limit can be approached. Since we do, indeed, believe the 15 nm mentioned in our title is feasible, we are claiming that it is now possible to build an ion microprobe having a resolution well below that available from radioautography.

2) The absence of radioactivity permits high concentrations of tracer isotopes without much danger of artifacts or toxicity. It seems worth emphasizing that radioautography sometimes involves significant radiation damage to the specimen even before it gets to a microscope.

3) Several isotopes giving comparable resolutions can be distinguished when used simultaneously. It is difficult to see how radioautography might accomplish this.

4) For all of the elements that have a high concentration in biological material (H, C, N and O) and many other elements, there are stable isotopes which are rare enough to be of interest as tracers. If the natural abundance of a potential tracer is high enough to cause a significant background, one could introduce the tracer into material depleted in the tracer isotope. The possibilities for radioautography are more limited.

5) There is no need to wait for the decay of a radioactive isotope before the locations of the tracers can be determined.

6) Very irregular surfaces can be examined just as in an SEM, whereas rough surfaces are a serious problem for radioautography.

The most important advantage is probably the potential high resolution. In order to approach this with lenses conveniently available, very small apertures must be used. This causes no diffraction problems with ions, but there is an intensity problem. We have attacked this problem in a number of ways, the most important being the development of a new source.^{7, 8}

The source we have developed (called an EHD source) consists of a cone⁹ of liquid metal, formed in a strong electrostatic field, which emits ions from the tip where the field is strongest. Our studies of the ion beam from a cone of gallium, together with a straightforward calculation of the performance of a proposed ion-optical system, indicate that, at an ion energy of 100 keV, we should be able to get a primary ion current of 6×10^{-11} A into a beam spot having a diameter of 15 nm. This calculation depended upon the measured energy spread⁷ of the gallium ions in a small solid angle on the axis of the cone (12 eV, FWHM); an upper limit⁸ of 50 nm for the effective size of the ion source; and a brightness of 1.4×10^6 A cm⁻² sr⁻¹ for the 21-keV ions which were within a half angle of 4×10^{-4} rad at the source.

One of the most serious problems associated with the development of a practical instrument will be learning to operate the source at its maximum brightness reliably and reproducibly. In addition, if no way is found to reduce the tendency of the image to move every few seconds, special electronic techniques may be needed in order to insure that a correction for this motion is provided so that successive frames will be properly registered with respect to one another.

Recently we measured the yield of negative ions (secondary ions/primary ion) from gelatin and demonstrated that the negative ion yield could be enhanced by evaporating neutral cesium onto the sample during the ion bombardment.¹⁰ Some of the negative ion yields observed by Benninghoven, Jaspers, and Sichtermann¹¹ from amino acids (without the use of cesium) are larger than the yields we obtained from gelatin with cesium enhancement. Hence, we believe the calculations¹⁰ using our results provide a conservative estimate of the negative ion yields to be expected from biological samples in general. Our scanning microprobe is intended to be used in tracer experiments which would be rather like the use of tracers in radioautography. In a typical radioautography experiment, one examines an electron micrograph of the developed grains associated with the decay of the tracers, with the specimen superimposed in the same micrograph. Likewise, in the proposed ion microprobe the structures of interest themselves will be delineated by a scan using some abundant ion from the sample or, if no ion shows sufficient discrimination, an ion from a stain, which could be any material furnishing a distinctive ion, not necessarily one containing a heavy element as in electron microscopy. The scan of these abundant ions should be almost automatically in register with the scan of the tracer ions.

Since, in order to trace stable isotopes, one must usually resolve mass doublets such as $^{16}\text{O}^1\text{H}^-$ and $^{17}\text{O}^-$, one must collect secondary ions containing the stable isotopes of interest into the acceptance phase space of a mass spectrometer capable of resolving mass doublets. This

requires mass resolutions, $M/\Delta M$, of about 10,000 and tends to aggravate the intensity problem. However, we have demonstrated⁵ that it is possible to collect approximately 10% of the secondary ions into the acceptance phase space of such a spectrometer. We should note though that the mass spectrometer considered was the 100-inch instrument at Argonne⁶, and that this is a very expensive machine. With more practical machines the 10% may become 3%.

We have calculated that, if structures occupying 1% of the area of a section under observation contained 1 part in 40,000 of the tracer material and the remainder contained relatively little, a scan of 250 sec (covering $10^3 \mu\text{m}^2$ and removing a 15-nm layer by sputtering) would give 25 counts in the 1% area and about 250 (roughly 1 per sec from machine background) counts in all the rest of the area. With about 9% of the 275 counts associated with the 1% area it would be highly probable that the tracer was concentrated there.

Radiation damage is of course total in that several atomic layers are removed in the course of examining a specimen. Moreover, there is serious damage in a layer 10 to 50 nm deep in the remaining material because atoms of the sample are seriously displaced and gallium atoms are imbedded in the sample. We would propose that if successive layers are to be examined, a low voltage etch using say argon ions be used to expose a new surface sufficiently free from damage after each successive microprobe examination of a given area.

We have a scanning microprobe under construction which is designed with the intention of demonstrating the soundness of our expectations and to serve as a test bench for work on improvements of the components. It is not however a user-oriented machine designed to handle a large demand in a routine way.

REFERENCES

* Work performed under the auspices of the U. S. Energy Research and Development Administration, Division of Physical Research and the National Institutes of Health.

1. ANDERSEN, C. A. & J. R. HINTHORNE. 1972. Ion microprobe mass analyzer. *Science*, N. Y. 175: 853.
2. LIEBL, H. 1975. Ion probe microanalysis. *J. Phys. E: Sci. Instrum.* 8: 797.
3. KISIELESKI, W. & R. RINGO. 1975. Microscopic tracing of deuterium. *J. Microsc.* 104: 199.
4. An exception is ^{125}I for which a resolution of 50–60 nm was achieved by SALPETER, M. M. & M. SZABO. 1976. *J. Histochem. Cytochem.* 24: 1204.
5. KROHN, V. E. & G. R. RINGO. 1972. Secondary ion collection system for an ion microprobe of high mass resolution. *Rev. Sci. Instrum.* 43: 1771.
6. STEVENS, C., J. TERANDY, G. LOBELL, J. WOLFE, N. BEYER & R. LEWIS. 1960. In *Proceedings of the International Conference on Nuclidic Masses*, Hamilton, Ontario, edited by H. E. DUCKWORTH (University of Toronto Press, Toronto, 1960): 403.
7. KROHN, V. E. & G. R. RINGO. 1975. Ion source of high brightness using liquid metal. *Appl. Phys. Letters* 27: 479.
8. KROHN, V. E. & G. R. RINGO. 1976. Microprobe design using a liquid gallium ion source. *Int. J. Mass Spectrom. Ion Phys.* 22: 307.

9. TAYLOR, G. 1964. Disintegration of water drops. Proc. R. Soc. Lond. A280: 383.
10. KROHN, V. E. & G. R. RINGO. 1977. Some proposed improvements in the scanning ion microprobe. (to be published)
11. BENNINGHOVEN, A., D. JASPERS & W. SICHTERMANN. 1976. Secondary-ion emission of amino acids. Appl. Phys. 11: 35.