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MASTER

AUTHOR(S): C. Maggiore

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LOS ALAMOS SCIENTIFIC LABORATORY
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DATA ACQUISITION WITH A NUCLEAR MICROPROBE

C. J. Maggiore
Los Alamos Scientific Laboratory
P.O. Box 1663, E-10, MS 430
Los Alamos, New Mexico 87545

Abstract

Spatially resolved information from the near surfaces of materials can be obtained with a nuclear microprobe. The spatial resolution is determined by the optics of the instrument and radiation damage in the specimen. Two- and three-dimensional maps of elemental concentration may be obtained from the near surfaces of materials. Data are acquired by repeated scans of a constantly moving beam over the region of interest or by counting for a preset integrated charge at each specimen location.

Introduction

The purpose of a nuclear microprobe is to obtain spatially resolved information from a specimen containing some nonhomogeneity. The nonuniformity may be in any direction relative to the incident beam direction, but the problem of data acquisition is always the same--where is the nonuniformity, and how big is it? There are three analytic signals used with a nuclear microprobe: particle induced x-ray emission (PIXE), Rutherford backscattering (RBS), and nuclear reaction analysis (NRA). The signal or signals detected, the nature of the specimen, the desired information, and the available beam current determine the appropriate solution to the problem of data acquisition. Microprobes existing in the world today have unique solutions, usually dictated by the constraints of the locally existing hardware for nuclear physics data acquisition. This paper cannot review all the existing and proposed solutions[1], but it discusses the general constraints of the problem.

Data Rates

The total amount of data processed depends on the incident beam current and the beam-specimen interaction. Partitioning the data into individual picture elements, or pixels, is necessary to obtain the spatially resolved information. Therefore, considerations of beam current vs spot size and time/pixel vs resolution, concentration, and cross section determine how the data can be acquired and stored.

An ideal ion-optical system focuses some fraction of the accelerated beam into the final spot with the amount determined by the ratio of the phase space acceptance of the final lens to the total available phase space.

$$i_{probe} = \frac{(\epsilon_x \cdot \epsilon_y)_{probe}}{(\epsilon_x \cdot \epsilon_y)_{source}} \times i_{source} \qquad (1)$$

where i is the current and ϵ is normalized emittance. A more realistic upper bound on the beam current vs spot size may be obtained by including the effects of spherical and chromatic aberrations on the final spot. Assuming that the geometrical aberrations add in quadrature, one obtains

$$d^2 = d_o^2 + d_s^2 + d_c^2$$

$$d^2 = d_o^2 + \left(\frac{C_s \alpha^3}{2}\right)^2 + \left(2 C_c \alpha \frac{\Delta E}{E}\right)^2 \qquad (2)$$

where d_o is the spot diameter, assuming perfect optics, C_s and C_c are the spherical and chromatic aberration coefficients relative to the image point, α is the semidivergence in the focused beam, and $\Delta E/E$ is energy uncertainty in the beam. The results of the calculations for the Los Alamos Scientific Laboratory (LASL) microprobe with a superconducting solenoid final lens are shown in Fig. 1. Current densities of 1 nA/μm^2 are possible with this system. If beams with $\Delta E/E = 10^{-4}$ are available, this current density can be maintained to 1-μm^2 spot sizes. Quadrupole multiplets usually have a smaller phase space acceptance for the final lens with a consequent reduction in final current density.

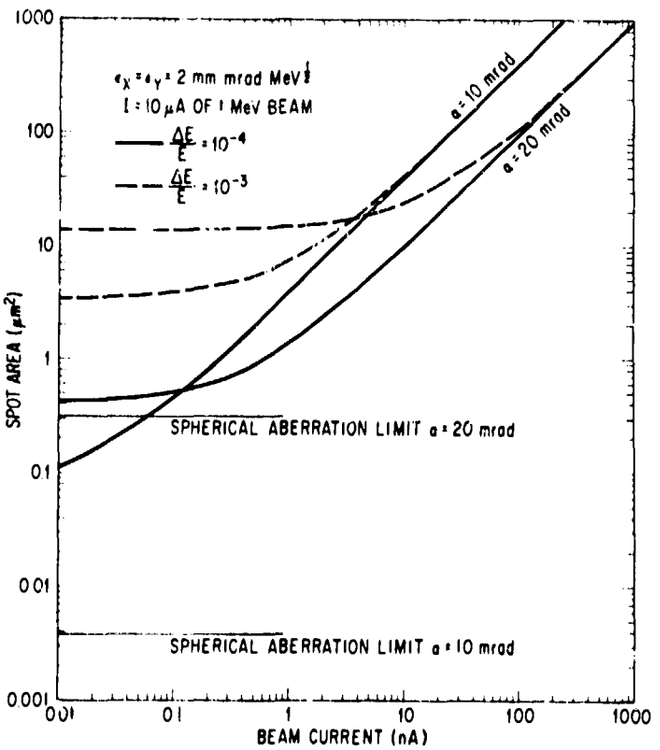

Figure 1. Beam current vs spot size, including the effects of spherical and chromatic aberrations for the LASL microprobe. The aberration coefficients C_s = 14 cm and C_c = 9.3 cm were calculated from the equivalent Glaser field $B(z) = B_o/(1 + (Z/a)^8)$ with a = 6.5 cm.

The time, τ, required to acquire the data from a single pixel depends on the incident beam current in particles/s, i; the number of target atoms/cm^2, Nt; the solid angle of the detector, Ω; the cross section for the interaction, σ; and the number of events to be counted, A. In general,

$$\tau = \frac{A}{i \Omega \sigma N t} \quad . \qquad (3)$$

A series of universal curves for the time/pixel vs the probe current is plotted in Fig. 2. The curves assume no background and should, therefore, be considered somewhat optimistic. Curves such as this are graphic reminders of the obvious tradeoffs between spatial resolution and sensitivity.

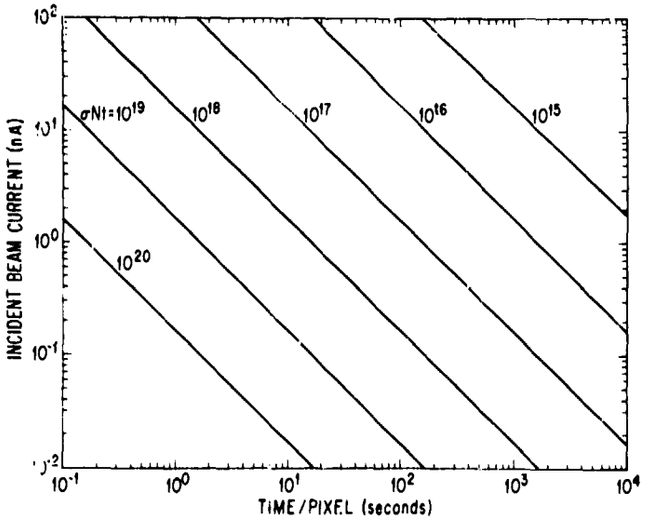

Figure 2. Time/pixel vs incident beam current for different values of concentration and cross section. σNt = barn atoms/cm². The time plotted refers to the time required to detect 100 counts with a detector of 10^{-3} str and 100% efficiency.

Radiation Damage

Since a nuclear microprobe is designed to tell you what elements are in the specimen and where they are, questions of elemental migration within the matrix are important. Thermal diffusion and nuclear recoils are two effects that move atoms in the specimen over the dimensions being measured. The following order of magnitude calculations are designed to tell whether you can get the information before the beam changes the parameter being measured.

An upper limit on thermal effects may be calculated by assuming uniform energy deposition in a cylindrical column of the specimen excited by the incident beam. Heat is radially conducted to the edge of the specimen. The solution to the radial part of the equation of conduction for heat supplied at a constant rate, F_o, per unit length on an inner cylinder of conductivity, K, is

$$2\pi K(T_1 - T_2) = F_o \ln\left(\frac{r_2}{r_1}\right) \quad . \tag{4}$$

Solving for T_1, the temperature at the edge of the excitation volume, yields

$$T_1 = T_2 + \frac{EI}{4\pi^2 r_1^2 RqK} \ln\left(\frac{r_2}{r_1}\right) \quad , \tag{5}$$

where E is the energy of the incident beam, I is the beam current, R is the range, q the charge on the incident particles, and r_1 is the radius of the incident beam.

Thermal conductivities vary over several orders of magnitude from 10^{-3} - 1 cal/cm s °C with most metals and semiconductors having K > 0.1 cal/cm s °C. For a 1-MeV beam of 10-μm projected range with beam current of 2 nA in a spot 1 μm in diameter, the temperature rise in a 1-cm specimen of thermal conductivity K = 0.1 cal/cm s °C is ~9°C. Localized specimen heating is probably not a problem in such specimens if the overall temperature rise is controlled by adequate thermal contact with a heat sink. For glass or biological materials with much smaller conductivities, localized heating can be a severe limitation. The data may have to be collected from a constantly moving point on the specimen to minimize thermal degradation. Data acquisition for a preset time or integrated charge at each pixel is possible with the more rugged specimens.

The question of nuclear recoil is intimately related to sensitivity and limits of detectability with a nuclear microprobe because the incident particle can transfer significant energy to the target nucleus. The kinematic factor for 180° scattering is

$$K = \frac{E'}{E} = \left(\frac{M - m}{M + m}\right)^2 \quad . \tag{6}$$

For incident alpha particles of a few MeV, the target nucleus will recoil in the forward direction with energies > 100 keV. The range of this recoil particle depends on its mass, energy, charge, and the matrix, but typically is ~0.1-1.0 μm. This displacement is greater than the depth resolution of RBS. Detecting the presence of an atom will change its location. It is not a problem with an unfocused beam, but it is one of the ultimate limitations for spatially resolved information.

A monolayer of heavy atoms (~10^{15} atoms/cm²) is easily detected with Q = 1 μC (6.25×10^{12} particles) of incident alpha particles, but within an area of 1 μm² there are only 10^3 atoms present. Six orders of magnitude more particles must pass through the monolayer to be detected than are present to be measured. As shown in Fig. 3, only the small fraction of particles scattered through large angles are of concern. The majority of incident particles scattered in the forward direction impart very little tangential momentum to the target nucleus, not enough to move the target atoms laterally out of the 1-μm² sampling area. The total number of particles scattered through an angle larger than Φ_{Min} is

$$A = QNt\left(\frac{z_1 z_2 e^2}{2E}\right)^2 \int_{\Phi_{Min}}^{\pi} \frac{\sin\Phi}{\sin^4\left(\frac{\Phi}{2}\right)}\, d\Phi \quad . \tag{7}$$

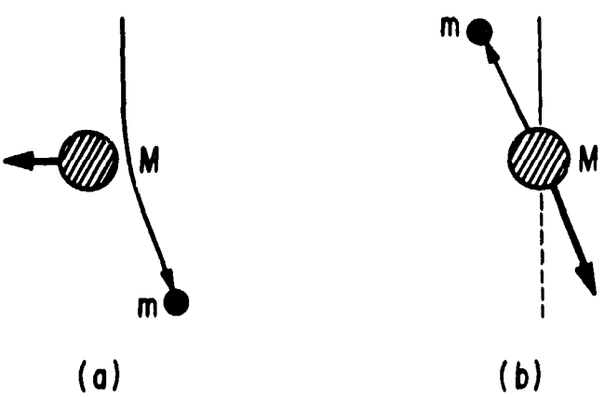

Figure 3. The majority of scattering events are forward scattering as in (a), where very little lateral momentum is transferred to the target nucleus, M. The backscattering events (b) move the target nucleus a distance greater than the depth resolution of the measurement.

For 1 μC of 1-MeV alpha particles on a monolayer of gold, 2 x 10^{9} incident particles will be scattered through a center of mass angle greater than 90°. The total detected count from such a layer would be ~200 counts with a 10^{-3} str detector at 175°. The process of making a ±7% measurement on 10^{7} atoms in 1 μm^2 would move ~2% of the atoms distances greater than the resolution of the measurement. There is no way of acquiring the data to avoid this fundamental limitation.

Information

The main strength of PIXE is an increased elemental sensitivity relative to electron-beam excited specimens for intermediate and heavy atoms. The energy dependence of the x-ray production cross section means that some elemental depth information can be obtained by differential measurements, but this is not easily applicable with a microprobe. The usual application of the PIXE signal from a nuclear microprobe is to obtain multielement two-dimensional distributions and use the increased elemental sensitivity at selected points of interest. The reason is readily apparent from Fig. 2 where σNt < 10^{16} barn atoms/cm^2 implies data acquisition times > 10^{3} s for a 1-nA beam current. It is not reasonable to think of a nuclear microprobe with PIXE as an instrument with 1-ppm sensitivity everywhere and with two-dimensional spatial resolution of a few μm^2.

However, in most applications, ppm sensitivity at every point is not required. The problem is to find those regions of interest requiring long counting times or localize other regions with concentrations greater than 10 ppm. Legge and Hammond[3] at Melbourne have taken a sophisticated event recording approach. The primary interest is the localization of heavy trace elements or impurities in thin biological specimens. The beam is magnetically deflected in a raster pattern on the specimen, and the XY positions and energy of each event are stored on tape or disk. A storage scope is used to display the data as two-dimensional elemental maps, line scans, point spectra, or selected area averages. The system works quite well because the count rates and total number of events to be recorded are low. The fast deflection of the beam minimizes thermal degradation of the specimen.

The RBS and NRA data acquisitions with a microprobe are complicated by an extra dimensionality of information, the depth distribution. The particles entering and leaving the specimen lose energy in a known way, and the distorted peak shapes contain the depth information. Figure 4 shows the spectrum obtained from 2-MeV deuterons on 250 nm of anodic oxide on GaAs. Both the RBS deuterons and the proton and alpha peaks from the nuclear reactions with ^{16}O are shown. The alpha particle peak at 2.9 MeV from the $^{16}O(d, \alpha)^{14}N$ reaction can be used to profile the oxygen concentration as a function of depth in the GaAs oxide.

To obtain the depth information, peak shape analysis must be performed at each pixel. Also, if the microprobe is used to obtain a two-dimensional array of such information, the data cannot be conveniently displayed because it is four dimensional--concentration and positions x,y, and z.

The obvious solution is to simplify the problem by idealizing the geometry. This usually means obtaining line-scan information rather than full area scans. Reducing the dimensionality of the problem is usually required for another reason, data retrieval rates. The cross sections for RBS and NRA are normally lower than for PIXE and require long data access times per pixel. Figure 5 is an example of such line-scan information showing the variation in oxygen concentration vs depth across a laser-annealed spot on GaAs anodic oxide.

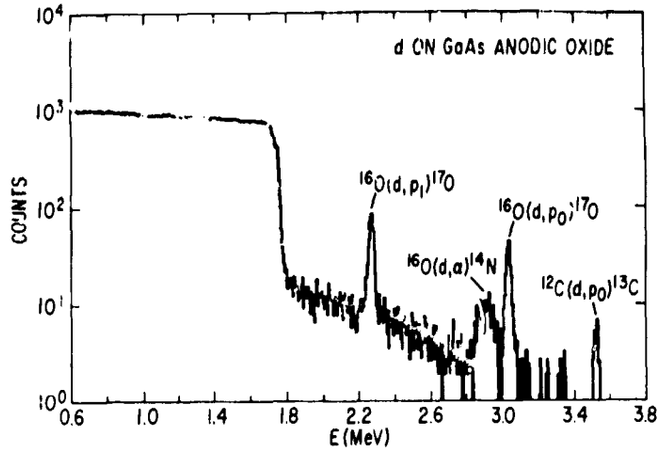

Figure 4. Backscattering and nuclear reaction spectrum of 2-MeV deuterons on a 250-nm GaAs anodic oxide.

Since the LASL microprobe[3] was intended to be used primarily for semiconductor and metallurgical applications where localized heating is not expected to be a problem, a fixed beam is used. Quantitative results are obtained by counting for a preset integrated charge at each pixel. Figure 6 shows the system with its computer generated XY rastering capability. In the usual mode of operation, complete spectra from the detectors are obtained at each point, and the information is written to magnetic tape for permanent storage and later analysis. Up to 72 gates can be set on the spectra, and the integrated sums or differences can be stored in memory. That is, 72 line scans derived from the spectra can be accumulated in memory during data acquisition. The complete spectral storage capability of this approach means that data retrieval rates are detector and beam current limited rather than computer limited.

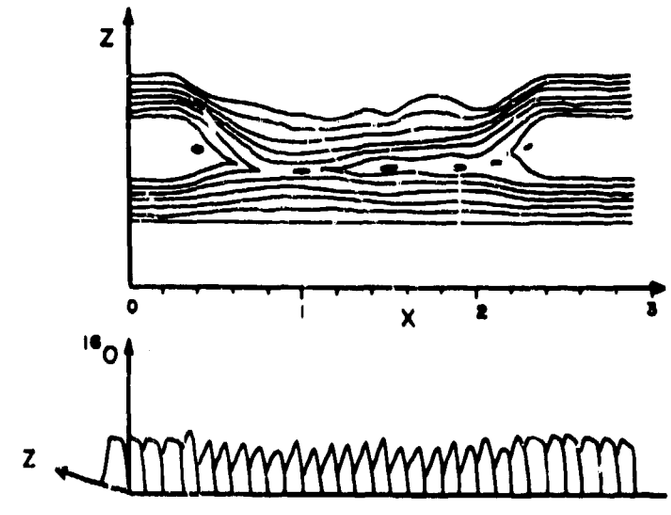

Figure 5. Contour and isometric plots of oxygen concentration vs depth in a 250-nm thick anodic oxide on GaAs. The oxide was annealed with a 70-ns pulse of 248.2-nm radiation from a KrF laser.

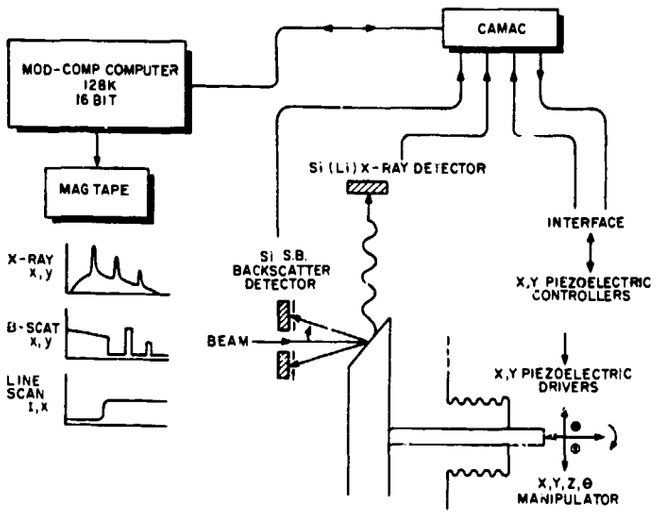

Figure 6. Data acquisition system for the LASL microprobe.

The optimal solution to the data acquisition problem is the one that records only the information required to answer the particular question about the sample. This means applying strong filters to the data before storage to reduce the mass of information. However, problems can arise with new or unknown samples where it is not obvious what filters to apply until after the data are acquired.

In addition to acquiring the analytic information from the specimen, the beam must be focused on the region of interest. This has usually been done with an optical microscope and thin scintillator, but the ability to image the specimen directly with the beam is a great convenience. The combination of a fast deflection system and specimen imaging from secondary electrons makes the nuclear microprobe similar to a scanning electron microscope[4]. Computer generated rasters and direct imaging will be a great help in making the nuclear microprobe a convenient instrument for precise, reproducible, quantitative information from the near surfaces of materials.

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