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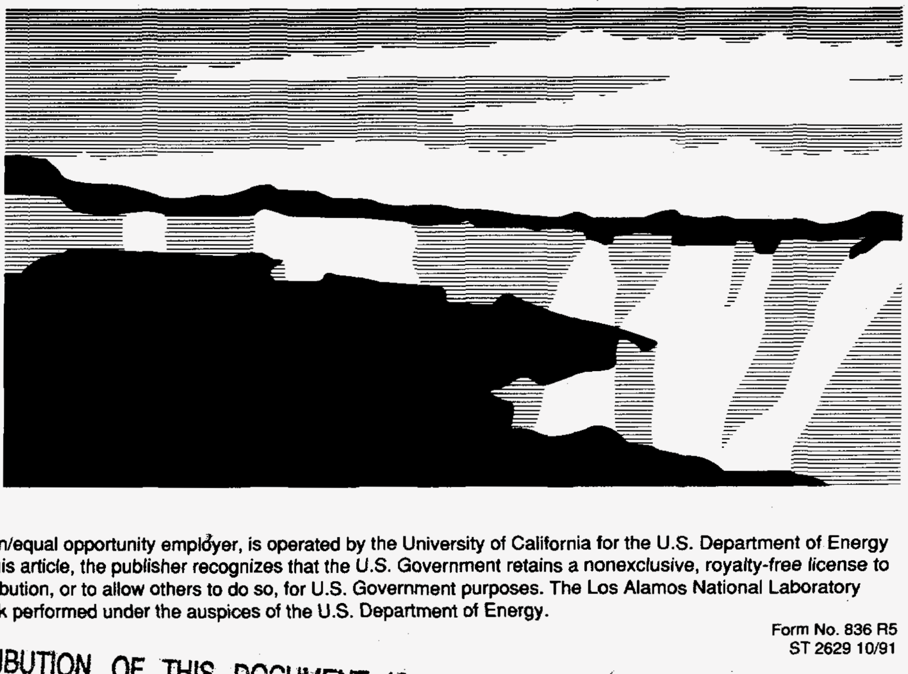

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SPALLATION SOURCES IN SUPPORT OF TECHNOLOGY

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ABSTRACT

In this contribution I summarise a number of recent experiments at the Los Alamos Neutron Science Center (LANSCE) that have contributed to strategic and applied research. A number of new tools have been developed to address these problems, including software that allows materials texture to be obtained during Rietveld refinement, Bragg-edge diffraction, resonant-neutron and proton radiography. These tools have the potential to impact basic as well as applied research. It is clear that a new, more powerful neutron source such as the planned Japanese Hadron Project will be able to use these and other techniques to contribute in a direct way to important industrial technologies.

1. Introduction

The neutron scattering community is justifiably proud of the contributions it has made to basic research in many areas of science. Information obtained using neutrons has contributed strongly to our basic understanding of phenomena in diverse systems of interest to physicists, chemists and biologists — think, for example, of how little we would know about excitations in quantum fluids, the spin-density-wave state of chromium, electronic back-donation in the bonding of organometallic compounds, or the conformation of proteins and DNA in nucleosomes without neutron scattering. However, illustrious as this history of neutron scattering may be, it is not the only type of contribution neutrons have made to our modern scientific and technological enterprise. Increasingly in recent years, we have witnessed the application of neutrons to later parts of the R&D cycle, to problems that have been called "strategic research" and even in areas that are "applied research" or "product development". The purpose of my talk at this meeting is to illustrate this aspect of research at spallation sources, using examples of work that has been done at the Los Alamos neutron Science Center (LANSCE). As you will see, some of this work is driven by the fact that our principal funding agency, the Office of Defense Programs within the U.S. Department of Energy, has a mission need to master the science behind certain technologies. Even so, most of the examples I have picked are equally relevant to the industrial sector and are appropriate fields of study for spallation sources that are not fortunate enough to have such a clear mission.

2. Structural Materials

2.1 Improved Welding Techniques

My first example is from a general area that has attracted attention at the majority of the world's spallation sources — residual stress. The failure of welds in beryllium is a long-standing problem. Processing conditions affect the failure of these welds and it is believed that the residual stresses in the region of the weld are sufficient to enhance cracking and failure. A step weld with an aluminium shim is being considered as a solution to this problem. It is expected that an interpenetrating structure of Be and Al will develop around the shim and that this composite material will be stronger than pure Be. Elastic strains in this region were studied by Mark Bourke and his collaborators at LANSCE with the weld under uniaxial compression, producing the results shown in Figure 1. Note that the data show strain perpendicular to the direction of the compressive force so that in a single-phase material one would expect to observe a tensile strain as a result of perpendicular expansion governed by Poisson's ratio. In deed, this is what is seen at low stress for both Be and Al. For higher stress, however, the aluminium clearly develops a compressive (not tensile) strain perpendicular to the load. Why is this? The detailed answer to this question is not yet known, but a model which includes the constraint on the aluminium imposed by the intercalated beryllium (which has a Poisson's ratio close to zero), gives the right trends. Clearly, without data of the type which neutrons can provide, one would have little chance of improving Be welds by design — trial and error would be almost the only recourse.

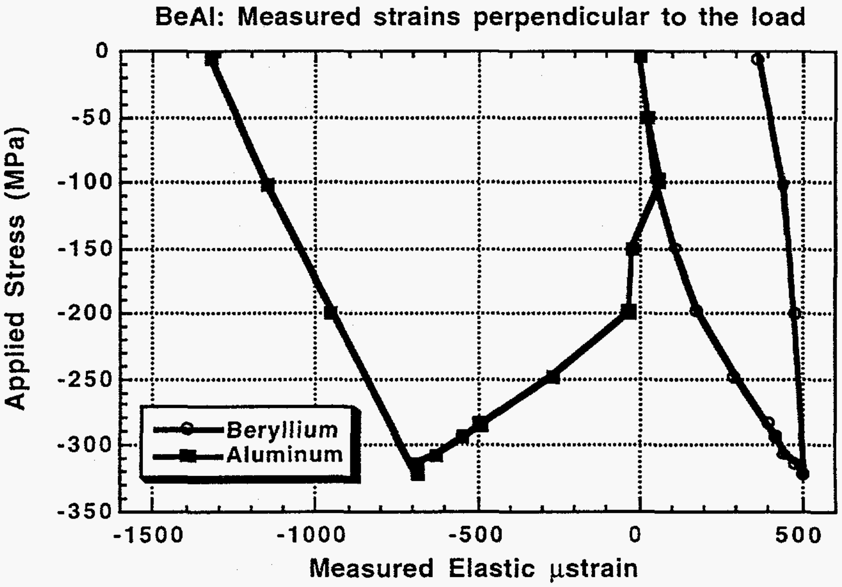


Figure 1: Elastic microstrain measured close to a beryllium-aluminium weld in a direction perpendicular to the applied compressive stress. The measurement was on the Neutron Powder Diffractometer at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC) at Los Alamos.

2.2 Texture

Another area in which neutron scattering is beginning to make significant contributions to applied research is in the measurement of texture — or preferred grain orientation — in polycrystalline materials. An important step in enhancing the tools for this type of measurement has recently been made by Bob Von Dreele at LANSCE. Bob has succeeded in extending the GSAS code (the Generalized Structure Analysis code that he and Allan Larson wrote to obtain structural parameters from diffraction data) to enable texture to be extracted as part of Rietveld refinement. Thus, pole figures no longer involve the measurement of a single Bragg peak at many sample orientations, but can be obtained from all of the peaks at once, even if many of them partially overlap their neighbours. Not only does this permit much faster measurement, but it also opens up the possibility of measuring

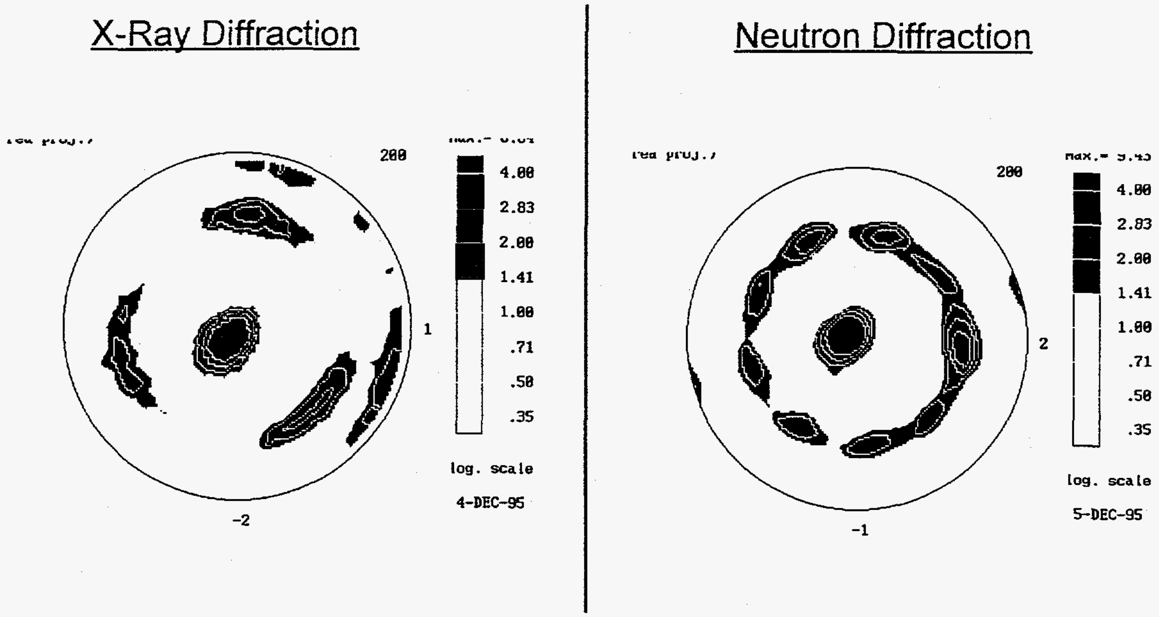


Figure 2: X-ray and neutron pole figures of upset-forged tantalum showing the difference between the surface and bulk textures.

texture easily in materials, such as high temperature superconductors, with complex crystal structures.

There are many examples of successful texture measurements at LANSCE, ranging from comparison of the textures of deep-drawn, spin-formed and cold-rolled uranium, to a study of textured titanium matrix compounds that will be used in the engine of the Advanced Tactical Fighter aircraft to reduce its weight by over 40 pounds. In the former case, neutron scattering showed that a proposed, low-waste process for forming uranium produced similar texture to a less benign manufacturing protocol, while in the latter it demonstrated that the texture of Ti wires was retained when the wires were rolled together to form a plate. In both of these cases, retention of a particular texture during processing is key to achieving desired mechanical performance.

In a very detailed study of tantalum processed by a technique known as upset forging, neutron scattering was able to obtain texture information which, when combined with known elastic constants, allowed a Los Alamos computer code (known as the Los Alamos Polycrystal Plasticity code) to predict the form of the anisotropic yield stress. The development of texture during processing could be followed for this high-temperature corrosion-resistant metal, and correlated with improvements in hardness. One point which emerged clearly from this study was the advantage of the neutron's ability to penetrate and probe the structure of bulk materials. Figure 2 shows x-ray and neutron pole figures for the (001) direction. The x-ray figure clearly shows only three (111) directions while the corresponding neutron results have 9 peaks of this sort, indicating that the average bulk texture is not the same as the surface texture. In fact, our texture program at Los Alamos uses neutron, x-ray, and electron diffraction as complementary probes to sample bulk, surface and local texture in a way that gives a much more complete picture than any of these techniques could give alone.

2.3 Phase Transformations

Texture is one property of materials which can strongly affect performance in real-world applications. The presence of various material phases is another. At LANSCE, we have developed a method for following phase composition which makes use of a technique called Bragg-edge diffraction. The method involves a measurement of the time-of-flight spectrum of neutrons transmitted through a polycrystalline sample using a current-mode neutron detector. In the spectrum are occasional discontinuities or dips that arise because neutrons on one side of the dip have wavelengths short enough to suffer a particular Bragg reflection, while neutrons on the other side have wavelengths that are too long for this process. Of course, there is a one-to-one correspondence between a Bragg-edge spectrum and a traditional diffraction pattern measured using scattered neutrons. The resolution of the Bragg-edge pattern is as good as that of diffraction in back-scattering geometry and the intrinsic signal is high because neutrons that are Bragg scattered in all direction contribute to a dip in the measured transmission spectrum. An important disadvantage of the method is that the background is intrinsically high so the signal-to-noise ratio is poor.

Using the Bragg-edge technique, Dallas Masters, a summer student at LANSCE, followed the reduction in the concentration of deformation-induced martensite in coupons of 304 stainless steel at high temperatures. One-minute measurements made at three minute intervals showed that more than 80% of the total transformation occurred during the first 15 minutes when the sample was heated to 670 °C or during the first 30 minutes at 570 °C. The importance of this result lies in the fact that deformation-induced martensite is responsible for stress-corrosion cracking but that in high temperature applications, the disappearance of martensite leads to so-called “healing”. It’s a good idea to understand whether a stainless steel aircraft-engine manifold might crack at 35,000 feet or whether it is going to heal before it cracks.

3. High Explosives

Although one-minute measurements with neutrons represents fairly rapid data collection compared with many traditional measurements using this technique, recent resonant radiography experiments performed at LANSCE are even quicker by several orders of magnitude. In this case, the resonance was observed during a single neutron pulse, while a high explosive was detonated, to observe temperature distributions and particle velocities associated with the explosion. This type of information, which is important for understanding both the performance and the safety of high explosives, has long eluded experts and is now being furnished using neutrons. In one recent experiment by Vincent Yuan and his collaborators, two pieces of high explosive sandwiching a 500 μm indium foil were detonated simultaneously as a neutron pulse passed through the specimen.

Clear Doppler broadening of the 9.07 eV resonance in indium was observed (cf Figure 3) as a result of the high temperature generated during the explosion. Indeed, with the high peak neutron intensity provided by the Manuel Lujan Jr. Neutron Scattering Center (MLNSC), temperatures can be measured with an accuracy of about 30 ° using this method. In another similar measurement, a tantalum foil was blown towards the neutron source by an explosion, allowing its 4.28 eV resonance to be observed twice in the time-of-flight spectrum — once immediately before the explosion and once, Doppler shifted, after the explosion (cf Figure 4). From the interval between the explosion and the second observation, the velocity of the tantalum — about 3.9 km/sec — could be deduced. Quite apart from the technological importance of measurements like this to the explosives industry, they open up a whole new domain of fundamental shock wave physics for study.

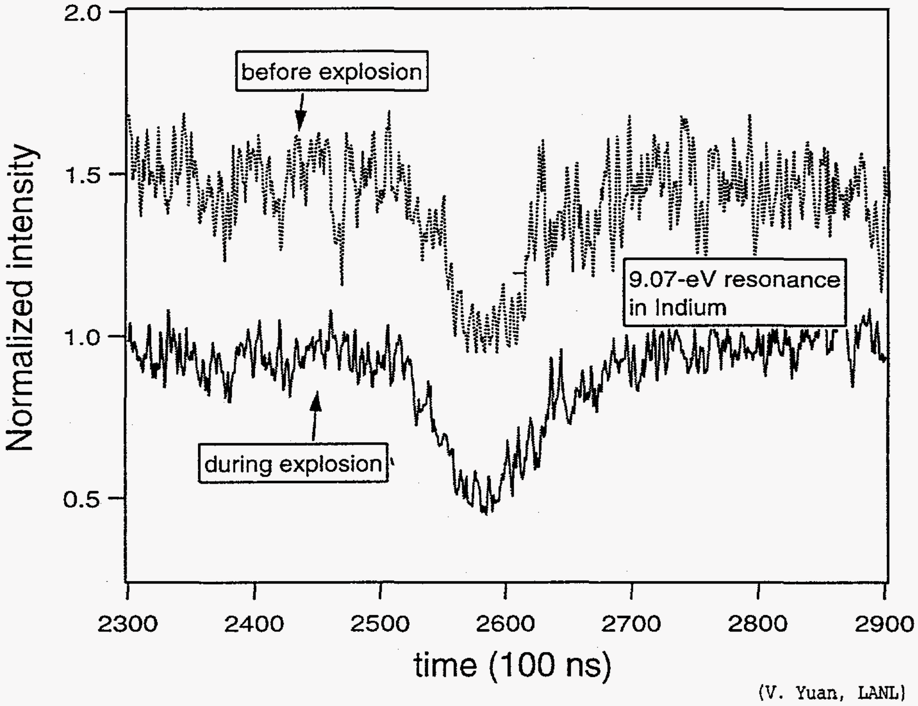


Figure 3: Neutron time-of-flight spectra obtained with a 500 μm indium foil during a single neutron pulse, before and during an explosion that compresses the foil. The broadening of the resonance recorded during the explosion is evident to the naked eye.

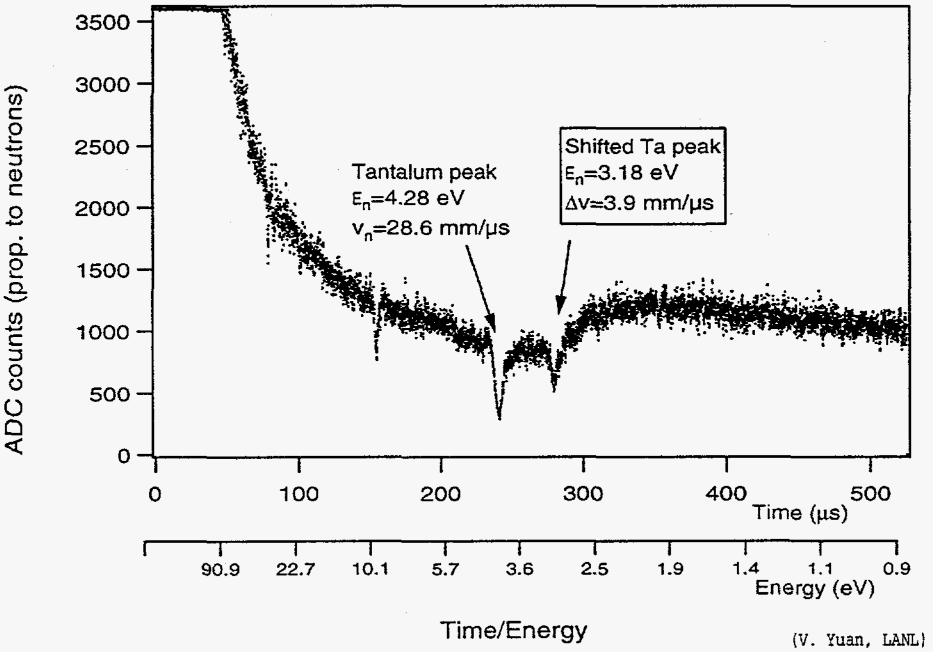
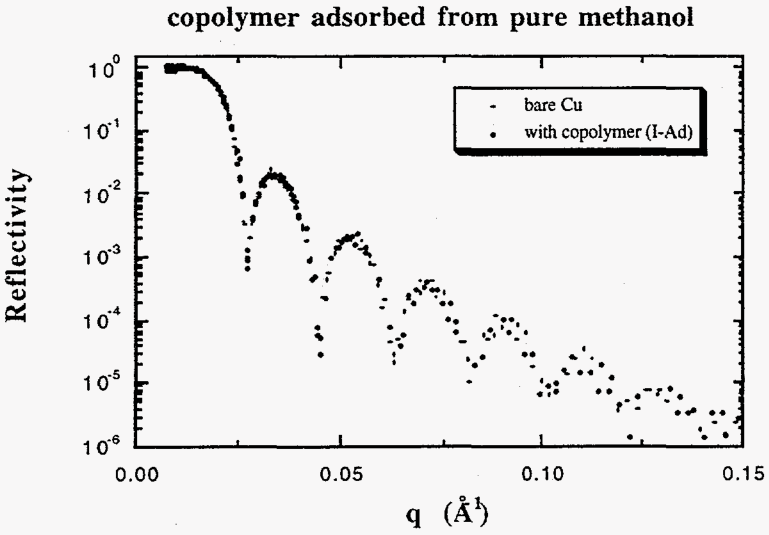


Figure 4: Neutron time-of-flight spectrum recorded during an explosion that forces a tantalum foil towards the neutron source. The 4.28 eV resonance is observed twice, allowing the velocity of the tantalum to be deduced.

In fact, as I show later, resonance radiography is one of the new tools being developed at LANSCE to support its defense mission that is likely to have future impact in basic science as well.

4. Better Adhesives

It is well known that neutron reflectometry has had an impact on our fundamental understanding of physical processes at surfaces and interfaces in a variety of systems. Recent experiments by Greg Smith at LANSCE and Mike Kent from Sandia National Laboratory have had a more applied motive however. In this case, the issue is to design a better adhesive to hold copper on to epoxy circuit boards. An obvious strategy is to use a polymeric adhesive that is composed of two distinct blocks — one which binds to epoxy and the other which binds to copper. Imidiazole / amine is a candidate. Previous work with this material had shown disappointing results when a good solvent for both blocks (methanol) was used. As the reflectivity curve in Figure 5 confirms, there is little difference between the reflectivity of a copper film deposited on a silicon wafer and that of the same sample after it has been dipped in the block-copolymer solution. For some reason, very little of the polymer is adhering to the surface, which is not good news for the circuit board manufacturers. However, when a poor solvent (methanol plus water) is used, the story changes, as Figure 6 shows. In this case, the raw data shows a substantial difference between the reflectivity of the dipped and undipped samples, and a model fitted to the data reveals a strong preferential adsorption of the imidiazole block to the copper surface, just as required.



copolymer adsorbed from pure methanol

Figure 5: Neutron reflectivities of a copper film on silicon and of the same system after it has been dipped in a block-copolymer solution that uses a good solvent. There is essentially no difference between the two measured curves.

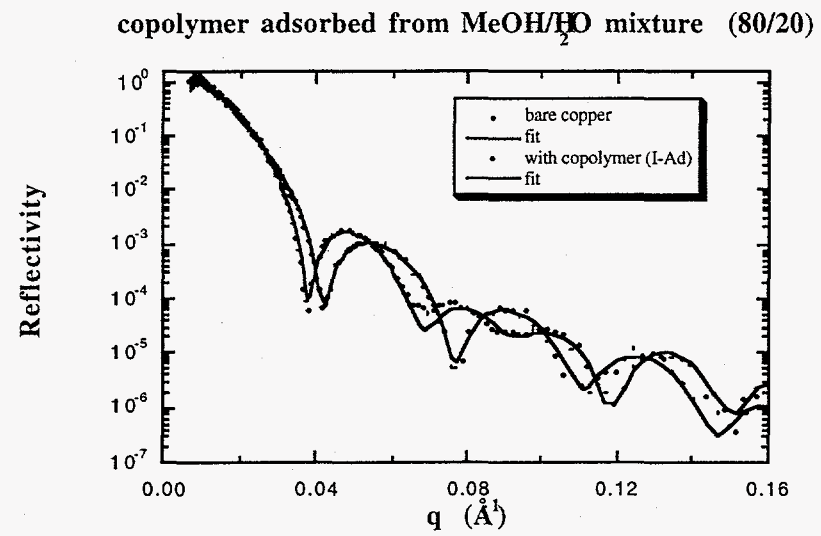


Figure 6: A repeat of the experiment shown in Figure 5 using a poor solvent for the block copolymer. The difference between the measured curves indicates strong adhesion of the polymer to the copper film. A model with one polymer block anchored to the copper gives a good fit to the data, as shown by the lines in the figure.

5. Single Event Latch-ups and Upsets

In addition to experiments that use neutron or proton beams to characterise samples, scientists at LANSCE have also been using the beams to modify samples in important ways. For example, together with colleagues from IBM, they have investigated the effect of proton irradiation and the resulting production of spallation products on the critical-current of high temperature superconductors and found a substantial enhancement. They have also made use of the fact that the neutron spectrum of the Weapons Neutron Research (WNR) facility — a spallation facility with a bare spallation target — is very similar to that produced in high flying aircraft by the interaction of cosmic rays with the airframe. The principal difference between the WNR neutron production and that at altitude is in the flux of neutrons produced, which is about 10000 times more intense at WNR. This large factor has enabled scientists from Boeing to test the effects of neutrons on aircraft electronics which, as feature sizes are reduced, may suffer from single-event latch-ups or upsets. Without knowing anything about these phenomena one can tell from the words used to describe them that they are not the sort of thing one wants to experience while flying over the Pacific ocean.

6. Proton Radiography

Finally, I want to mention a technique whose development has just started at LANSCE, proton radiography. At high enough energies, the principal interaction between protons and matter is very similar to the interaction between neutrons and matter, so one might imagine that high energy protons could be used for radiography. The problem is, of course, that protons suffer from multiple scattering. However, by using a system of quadrupolar lenses, it is possible to refocus a beam that has passed through an object and image features within the object with millimeter-level resolution. There is a limit to how thick the object can be because protons also lose energy when they pass through matter, causing chromatic aberration in the lenses, which gets worse at low proton energies.

Nevertheless, this new technique seems to have great promise for measuring both the density and type of material in an object. The latter information can be obtained because the degree of multiple scattering depends on atomic charge, Z. By choosing a particular aperture through which to create a radiographic image, one is therefore able to choose essentially the maximum value of Z that is imaged. Of course, protons have the distinct advantage over neutrons that their charge can be used as a handle to manipulate trajectories, allowing, in principal, multiple simultaneous images to be recorded from different angles.

This powerful new technique is being applied to problems of interest to the Office of Defense Programs. However, as it is developed, I expect to see its applications to problems in both basic research and the industrial sector, a healthy synergism between the various applications of beams at a spallation source.

7. Conclusion

I have tried to give a flavour of some of the more direct applications of neutron and proton beams to strategic and applied research. As more powerful spallation sources are brought on line, I expect to see even more such applications, especially if the sources are operated more or less continuously during the year and if we can invent ways in which industries can obtain the rapid access they need to be able to use neutrons to impact their ever shorter product development cycles. An important challenge will be to find ways to reduce the cost of obtaining the information companies need. One way to do this will be by increasing the neutron flux and the number of detector pixels so that information can be obtained quickly. This way, the fraction of a facility operating budget consumed to obtain a given piece of information — the texture of a new engineered component, for example — will be reduced. In this sense, a new source like the proposed Japanese Hadron Project can be expected to have significant industrial impact.