

A comparison of germanium and copper analyzers for pulsed-source crystal-analyzer spectrometers

M. Yethiraj and R. A. Robinson
 Los Alamos Neutron Scattering Center
 Los Alamos National Laboratory
 Los Alamos, New Mexico 87545
 USA

ABSTRACT: We describe the new copper "organ-pipe" analyzer installed on the Los Alamos Constant-Q Spectrometer^[1] and compare its performance with the previous germanium analyzer. In addition, we discuss the implications of our experience for pulsed-source crystal-analyzer spectrometers in general.

1. Introduction

It has long been realized^[2] that, for thermal and hot neutrons, beryllium is the material of choice for crystal monochromators and analyzers. On the basis of reflectivities, copper and silicon are next best, but the silicon coherent scattering cross-section is so small that silicon monochromators and analyzers would be impractically large (in thickness)^[2]. Given that it is only recently that single crystal beryllium has been grown reproducibly with sufficient quality for neutron monochromators^[3], copper is still the most practical choice for most thermal and hot neutron applications. On the other hand, if second-order contamination is a problem, the odd-index reflections (like (111), (113), (331) etc.) of a diamond-structure material like germanium can be used. The second-order reflections are systematically absent. However, a significant price will be paid in reflectivity (and hence intensity at the detector) when compared with a copper monochromator or analyzer. Within the neutron scattering community, a folklore has built up that order contamination is a significant problem on crystal-analyzer spectrometers and that one must therefore use germanium as an analyzer, even though the orders of reflection are separated by normal time-of-flight analysis. It seems that this belief has its origins in early experiments^[4,5] in which the (004) or (006) reflections of pyrolytic graphite were used. It is straightforward^[6,1] to show that, for an analyzing energy E_F and a primary to secondary flight path ratio L_I/L_F , the limiting condition for observing an excitation of energy transfer E in the presence of n th-order elastic contamination is:

$$\frac{E}{E_F} \leq \left[\frac{n}{1 - (n-1)(L_I/L_F)} \right]^2 - 1 \quad (1)$$

While this is not the whole story, in that the corresponding n th-order energy gain processes (from thermally populated phonons or magnons) and the effects of finite resolution are not included, it does provide a reasonable estimate of the constraints imposed by order contamination. For second order contamination ($n = 2$) and $L_A/L_F = 3.5$, which is roughly what we now have on the Los Alamos Constant- Q Spectrometer, this gives $E/E_F \leq 6.9$. Since one would not normally consider transferring much more than half of the incident energy to the sample, second-order contamination should not be a problem. Together with the fact that we had already done successful experiments with the germanium (220) reflection, which does have second-order contamination, this line of argument led us back to copper as an analyzer material

2. The analyzer

The "organ-pipe" analyzer geometry employed on the Los Alamos Constant- Q Spectrometer has been described previously^[1] and is illustrated schematically in Fig. 1. It has three advantages over the conventional disc geometry:

1. the combination of (a) having the cylinder axes parallel to the $[1\bar{1}0]$ axis and (b) pressing the original single crystal blocks parallel to the $[110]$ axis, as described in Ref. [1], means that one has a large horizontal and narrow vertical mosaic spread for all reflections of the type $(h h k)$.
2. the whole analyzer is used all of the time, in contrast to the disc geometry. This means that the minimum amount of material is used.
3. the analyzer thickness can be varied as a function of scattering angle, so as to optimize the reflectivity, as shown in Fig. 1.

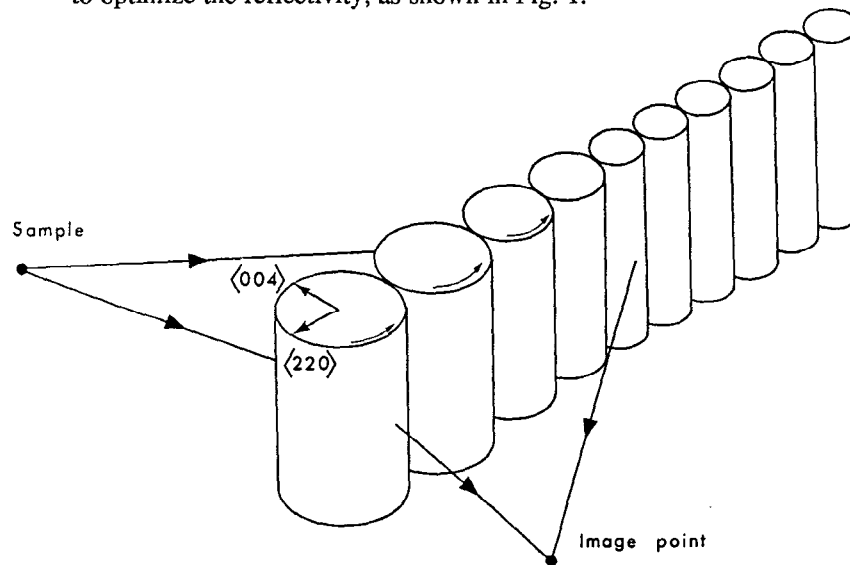


Fig.1 A schematic figure showing the "organ-pipe" geometry.

The photograph in Fig. 2a shows our original germanium analyzer. The original oriented germanium single crystals, of size 4 x 4 x 8 cm were bought from Eagle Picher Industries and hot-pressed in situ on a γ -ray diffractometer at the Institut Laue Langevin, Grenoble. The mosaic spread is approximately 30'FWHM. Unfortunately, the original single crystals had significant small-angle grain boundaries, as detected in γ -ray scans and the boundaries widened in angle during deformation. Some of the mosaic spread is therefore non-uniform. The germanium cylinders were then diamond core-drilled from the rectangular blocks used for pressing. These crystals were then oriented on a two-axis diffractometer at the University of Missouri Research Reactor and glued, with dental cement, on to the mount shown in Fig.2a. This consists of a set of vertical shafts driven by a worm and gear system from two horizontal shafts geared to a stepping motor. We wished to locate the crystals as close together as possible and this necessitated the use of two horizontal shafts rather than one. This system has worked well. The only problem encountered with it was that one of the horizontal shafts slipped, along its axis, relative to the other, with the consequence that the even crystals were misoriented by about 1° with respect to the odd crystals. We believe that this was due to incorrect seating of one of the bearings during assembly. This manifested itself in the form of double peaks in scans with an elastic incoherent elastic scatterer and the system was realigned on a two-axis diffractometer at the Omega West Reactor at Los Alamos.

Figure 2b shows the copper analyzer, which was constructed in a similar fashion. Again, plastic deformation was performed in Grenoble, cylinders were spark-eroded from the original boules and the analyzer was assembled on a two-axis neutron diffractometer at Missouri. In this case, we found that the optimum thickness varied much less strongly than for germanium. We therefore decided to use a single diameter (15 mm) for the cylinders. This gives us more flexibility in positioning the analyzer, as it is no longer optimized for specific scattering angles. In addition, optical encoders were installed on the horizontal shafts to aid in diagnosis of problems like that described above. The mosaic spread was approximately 20' FWHM.

3. Results

For comparison purposes, we show results for the elastic line from a standard ZrH_2 sample with both the germanium (331) and the copper (220) reflections. The d-spacings for these reflections are 1.2979 Å and 1.2780 Å respectively. Sections of representative scans are shown in Fig. 3. Figure 4 shows the variation of integrated intensity as a function of scattering angle within our spectrometer. The normalized integrated intensity is about 4.5 times greater for the copper analyzer. This is a much greater difference than the 70% increase calculated using the program MONO^[7]. In addition, the lineshape (see Fig.3) is much cleaner. This is due to a more ideal mosaic spread in the case of the copper analyzer. There was no significant difference in the background levels with the two analyzers.

4. Discussion

Returning to Eqn. 1, one is very unlikely to build a spectrometer with L_F outside the limits $0 < L_F < L_I$. The corresponding energy ratios, E/E_F , lie between 3 and

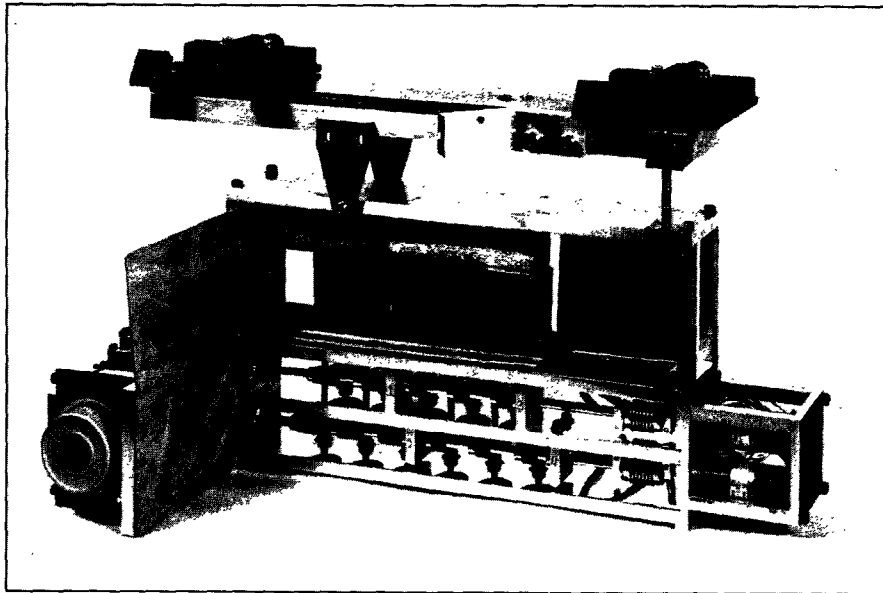
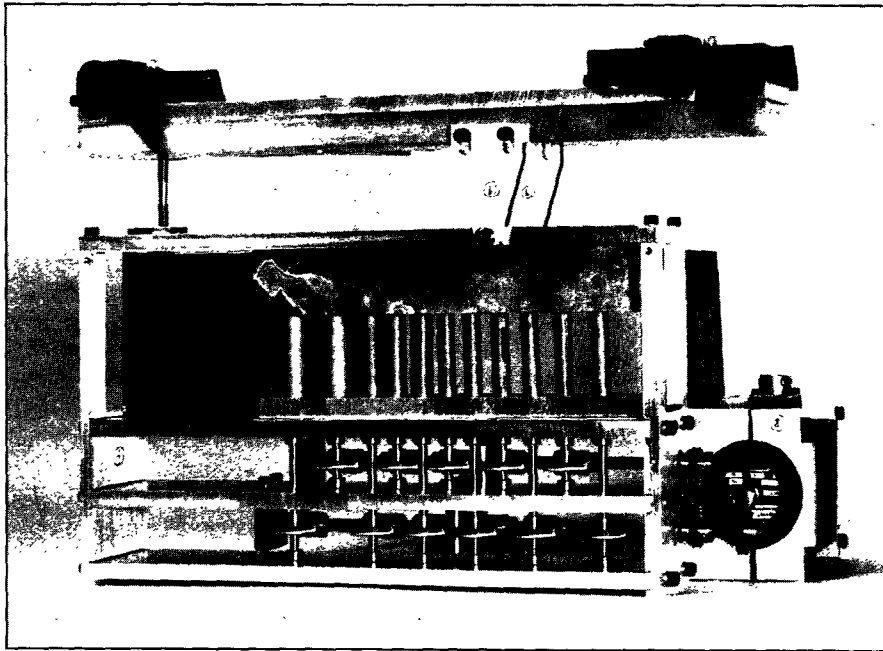


Fig.2 Photographs of (a) the germanium analyzer and (b) the copper analyzer.

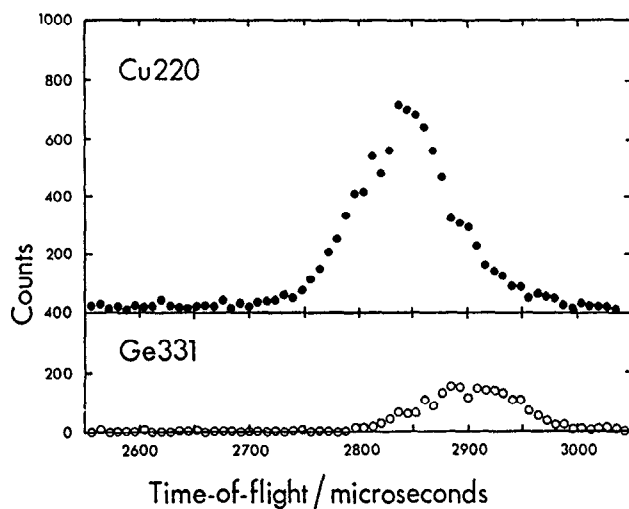


Fig.3 A typical time-of-flight scan through the incoherent elastic peak from ZrH_2 for Cu(220) and Ge(331) analyzers. The d-spacings differ by less than 2% and the monitor count was approximately 10% greater for the germanium run.

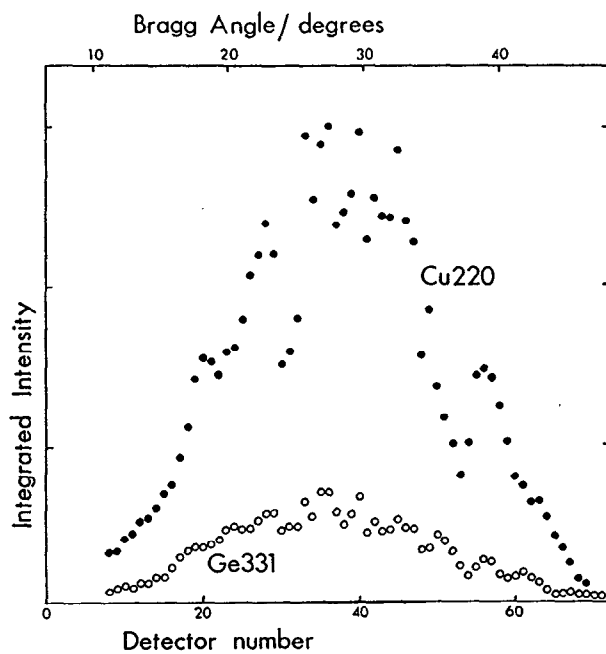


Fig.4 Integrated intensities (normalized to the same monitor count) for a wide range of detectors and the two analyzers Cu(220) and Ge (331).

infinity, for $n=2$. So, even in the worst case, second-order contamination should not be a problem, although it is fair to state that the longer the secondary flight path is, the less of a problem one has with order contamination. We conclude that there is no compelling reason to use diamond structure analyzer materials on pulsed-source crystal-analyzer spectrometers, unless one is consistently depositing most of the energy in the sample. One can always achieve a higher reflectivity with copper and the relative performance will be even better than one calculates. This is simply because it is easier to achieve good plastic deformation with the latter and, as a consequence, one is closer to the ideal mosaic model and the observed copper reflectivities will be nearer to the calculated values. Even on existing spectrometers with germanium analyzers it is simple to check the points raised above: use of reflections like (220), which are contaminated by second-order, should give more intensity than the odd-indexed reflections as the structure factor is twice as large.

Acknowledgements

We are indebted to A. K. Freund for numerous discussions and invaluable assistance in the fabrication of both analyzers. We are also grateful to the staffs of both the Institut Laue-Langevin and the University of Missouri Research Reactor for assistance in deforming and mounting the analyzer components. This work was supported, in part, by the division of Basic Energy Sciences of the US Department of Energy.

References

1. R. A. Robinson, R. Pynn and J. Eckert, 1985, Nucl. Inst. Meth. A241, 312.
2. A. K. Freund and J. B. Forsyth, 1979, in "Neutron Scattering in Materials Science", ed. G. Kostorz, Academic Press, p.477.
3. A. K. Freund, S. Jönsson, S. Stiltz and G. Petzow, 1984, J. Nucl. Mater. 124, 215.
4. C. G. Windsor, R. K. Heenan, B. C. Boland and D. F. R. Mildner, 1978, Nucl. Inst. Meth. 151, 477
5. R. Pynn and C. G. Windsor, private communication.
6. C. G. Windsor, 1981, "Pulsed Neutron Scattering", Taylor and Francis, London, ch. 9.
7. R. A. Robinson and A. K. Freund, 1985, Institut Laue-Langevin Report 85FR13T.