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TESTS OF A RESONANCE DETECTOR SPECTROMETER FOR ELECTRON-VOLT SPECTROSCOPY

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ABSTRACT

We have tested a resonance detector spectometer at the KENS neutron source, using ^{181}Ta, ^{121}Sb and ^{149}Sm absorbers and bismuth germanate (BGO), NaI and plastic scintillators. In the process we uncovered and solved numerous background problems, and discovered a time-focussing principle. We measured the scattering from a number of materials and so far have analyzed and present results for bismuth and graphite. Tests of cooled absorbers have indicated that resolution of 70 meV is possible with ^{181}Ta.

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1. INTRODUCTION

Spectroscopy using neutrons in the range 1-10 eV opens the prospect for several new kinds of measurements exploiting the short pulses and high intensity of epithermal neutrons produced by pulsed sources. Allen, Mitchell and Sinclair[1] have recently reviewed the principles and applications of these spectrometers, examples of which are in measurements of struck-particle momentum distributions, high frequency, low-wavevector excitations and molecular spectroscopy.

Heavy elements exhibit narrow nuclear resonances in the range of a few electron volts. These make possible spectrometers based on filter-difference methods or on detection of secondary capture products. We chose to develop a resonance detector spectrometer (RDS) based on detecting prompt capture gamma rays. This class of spectrometer gives the prospect of statistically cleaner results, as opposed to filter-difference spectrometers, particularly where the scattering function is small compared to its average value. Figure 1 schematically shows the resonance detector spectrometer. Scattered neutrons captured resonantly (at known energy E_f) by the absorber produce a cascade of gamma rays which register as pulses in the scintillation detector. The time-of-flight spectrum gives the incident energy dependence of the scattering probability.

(1) D. A. Allen, E. W. J. Mitchell and R. N. Sinclair, J. Phys. E: Sci. Instrum. 13 (1980) 639

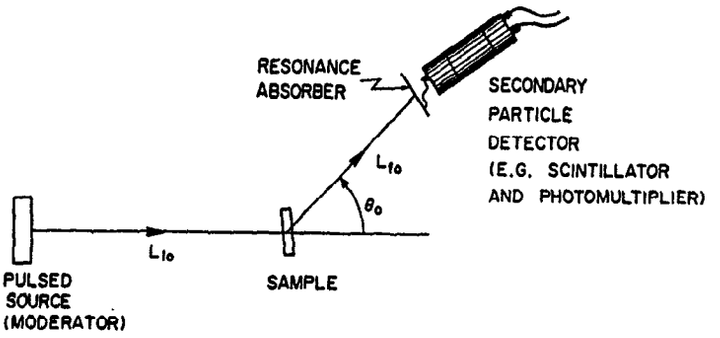


Fig. 1 Schematic diagram of a resonance detector spectrometer

The table shows some of the most-attractive resonances.

Table I Some Attractive Capture Resonances

Isotope	E_f, eV	Γ, meV
^{238}U	6.67	22.
^{181}Ta	4.28	57.
^{121}Sb	6.24	88.
^{149}Sm	.87	60.

We chose the 4.28 eV ^{181}Ta resonance for most of our tests because it has reasonably good resolution, lies in the range of energies of interest, and the material is readily available in appropriate thickness. Figure 2 shows the time-of-flight distribution of the capture gamma ray intensity from a 300 K, 12 μm foil of ^{181}Ta placed at the sample position of the spectrometer. The interval between 4.28 eV and the next-highest resonance at 10.34 eV is available for spectroscopy.

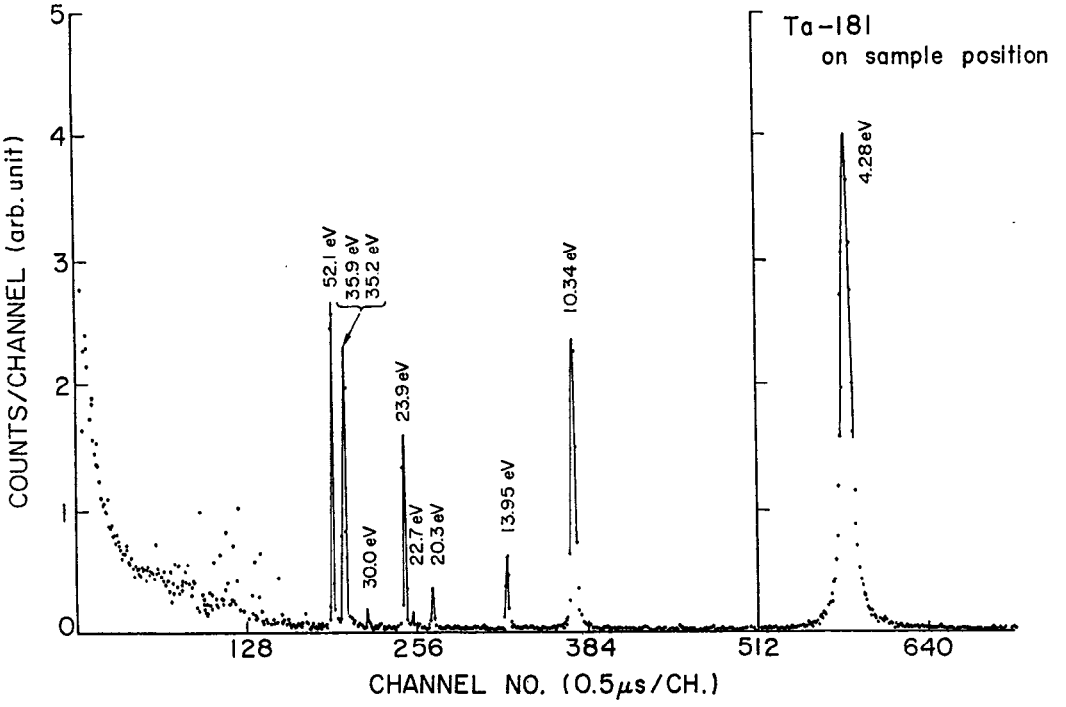


Fig. 2 Capture gamma ray intensity vs. neutron time-of-flight, for 12 μm Ta foil at the sample position (8.21 m, 0.5 μs/ch)

Figure 3 shows the 4.28 eV resonance in detail; the points are measurements, the solid line a first-principles calculation of the spectrometer response. The distance from moderator to absorber was 8.21 m for this test. The calculation includes the effect of geometry as well as self-shielding, Doppler broadening, and the intrinsic resonance width. We have performed a similar measurement and analysis of the 6.24 eV ^{121}Sb resonance and obtained similar agreement with a first principles calculation.

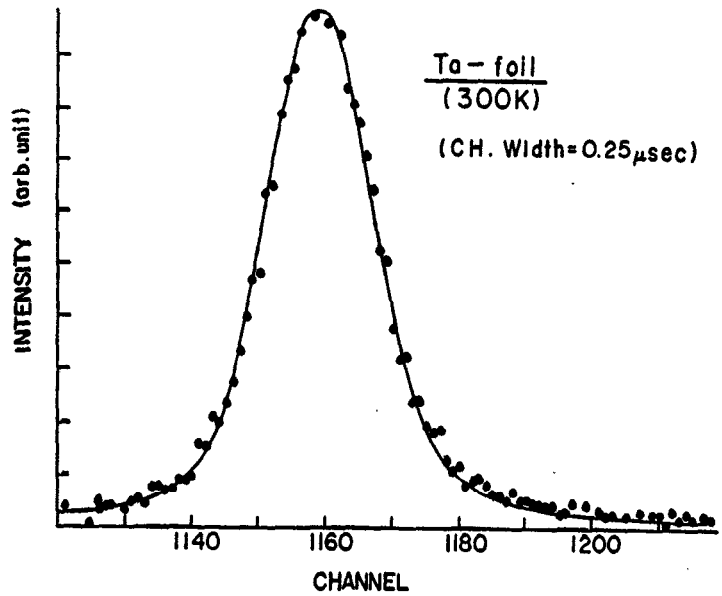


Fig. 3 The 4.28 eV ^{181}Ta resonance. Points, data of Fig. 2. Line, first-principles calculation, see text.

2. SHIELDING AND BACKGROUND

After extensive tests to identify sources of background and find corrective measures, we arrived at some general principles and some specific understandings which guided our development. Both neutron and gamma ray shielding emits capture gamma rays which can be detected by the scintillator. Both act as neutron traps, storing neutrons for several hundred microseconds. There is the possibility that some more-or-less short-lived (10-1,000 microseconds) isomers are produced in shielding and other components due to high energy neutron interactions, which decay during measuring time to produce detectable gamma rays. About 10 cm of lead is needed around the entire spectrometer to attenuate gamma rays from the surrounding concrete and steel. Beyond this, about 10 cm of hydrogenous material is needed to stop neutrons from outside. Polyethylene is inappropriate because of the 150 μs decay time of thermal neutrons in this medium; the thermal neutrons emit capture gamma rays (2.2 MeV) upon capture in hydrogen, and a 7 MeV cascade when they are captured in the adjacent lead. Boron loaded resin material works well.

B_4C shielding inside the spectrometer seems like a good idea. We tried configurations both with and without it, and at our levels of background, did not observe significant differences in the background. High energy neutrons accompany slow neutrons from the source, appearing in a difficult-to-stop halo around the beam. We finally found that very tight, massive collimation (lead, about 1 meter long, 40 cm dia.) around the indicent beam is required to deal with these neutrons. With this collimation, including B_4C and hydrogenous material, we were able to operate the detector within 10 cm of the center of the 4 cm wide beam.

We have tested various scintillators for gamma ray detection. Scintillator materials capture neutrons both resonantly and continuously; the resulting capture and decay gammas are detected with high efficiency. The traditional NaI detector is especially bad this way. We adopted bismuth germanate (BGO) ($Bi_4Ge_3O_{12}$) scintillators, which seem quite good in this application. Plastic scintillators have rather too low efficiency for the energetic gamma rays we must detect.

We measured the response of the BGO detector without a resonance absorber, and with a Pb scatterer. Most of the resonances are those of germanium. The spectrum is smooth, and the counting rate small for times longer than the arrival time of 40. eV neutrons.

Neutrons captured in the samples produce a sample-dependent capture gamma ray background in some cases. The vanadium 1/v cross section is large enough to be troublesome, giving a large constant background in the TOF spectrum. The vanadium sample contained a small amount of tantalum impurity, even though it is some of the highest-purity, zone-refined material. Since we were using the 4.28 eV resonance of tantalum as our monochromator, this interfered with measurement of the scattering. We made an antimony absorber, with which we satisfactorily measured the scattering from vanadium. The problem of 1/v capture in the important case of hydrogen is not so severe, since the ratio of scattering to capture is higher than in vanadium.

Some photomultiplier components contain materials having resonances in the neighborhood of those we want to use as monochromators. Gamma ray cascades from captures there are detected with high efficiency by the nearby scintillator, and may interfere with measurements as a structured background.

A persistent feature in the measured scattered neutron spectra was due to ^{121}Sb capture (6.24 eV). Since this moved (in time) according to detector position, we suspected it to be due to capture in some component of the detector assembly. We measured the capture gamma ray spectra of black dielectric tape, mu-metal and the photomultiplier dynode and photocathode regions, respectively used as light tight assembly material, magnetic shield and scintillation counter, irradiated in the sample position. Figure 4 shows the results.

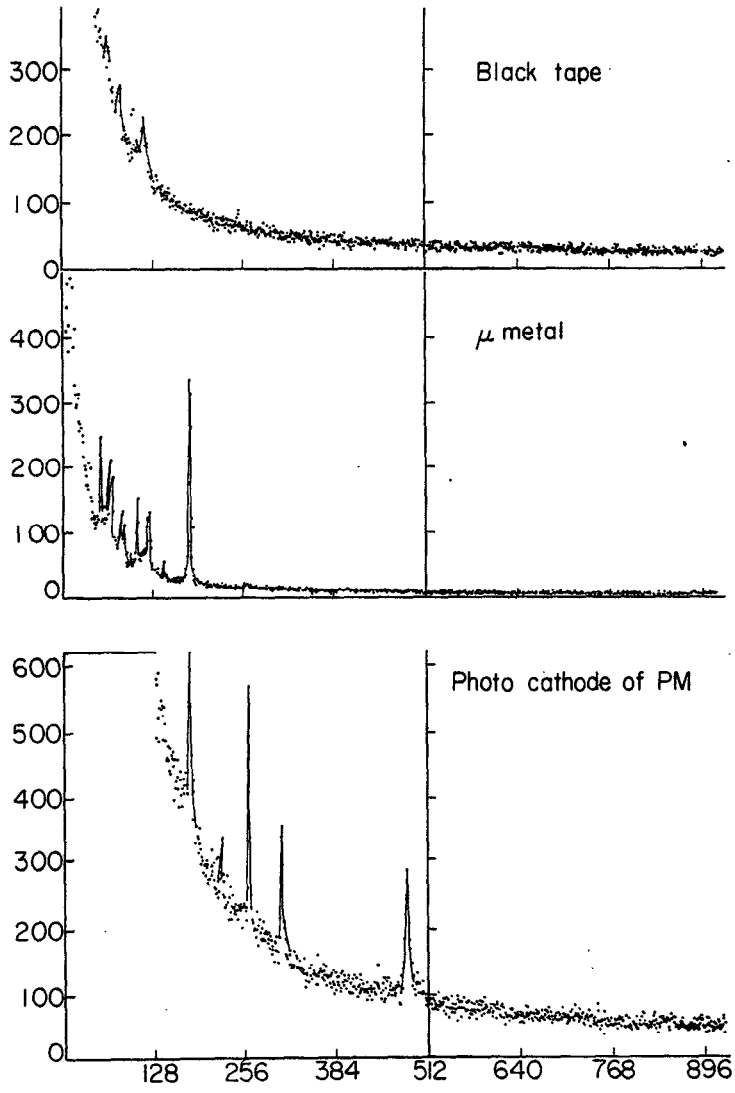


Fig. 4 Capture gamma ray intensity vs. time, for photomultiplier components irradiated at the sample position.

Even though the photocathode is so thin as to to be transparent, the troublesome 6.24 eV ^{121}Sb capture peak was evident only in the spectrum of the photocathode. Subsequently, we prepared a 1. mm thick shield of ^{10}B bound in epoxy resin; placed between the resonance absorber and the scintillator and photocathode, this reduced the spurious peak, and also reduced the background due to neutron capture in the scintillator.

Important in all this is that capture gammas appear after only about 10% of the captures in boron, moreover, their energy is low enough that we can electronically discriminate against them. Thus we have been able to freely use boron in the shielding.

3. RESOLUTION AND DEAD TIMES

The resolution accomplished so far is only modest, around 100 meV, limited by the fact that absorbers have been subject to room-temperature Doppler broadening, as well as by the lack of a uranium-238 absorber (which has the narrowest resonance we are aware of). Counting rates have enabled measurements to be completed in between a few hours and about ½ day.

We discovered a geometric focussing effect on the resolution, which comes about due to the joint effects of varying flight paths and scattering angles according to emission and interaction positions at the source, sample and detector. The recoil shift of the incident energy varies according to scattering angle, and times-of-flight vary according to incident energy and flight path length. The result is the subject of a separate paper.

We have examined a fast and a slow electronics for this application and found that the fast system worked well, having a dead time of about 100 nsec, while the slow one gave serious dead time problems in the TOF spectra.

4. SCATTERING MEASUREMENTS

We made test measurements on samples of Bi metal, V metal, graphite and H_2 gas. Figure 5 shows the results for 90° scattering from Bi, along with a

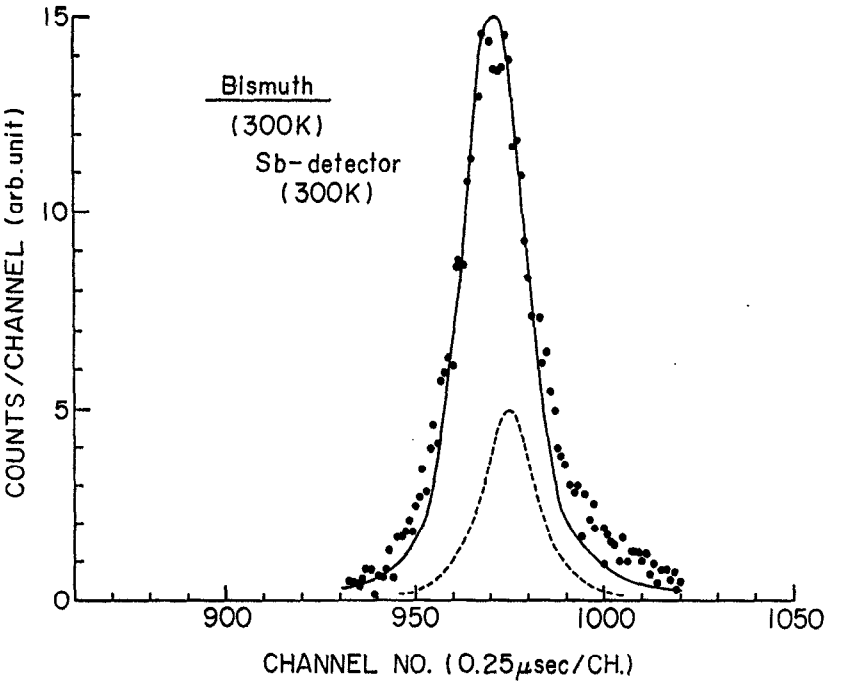
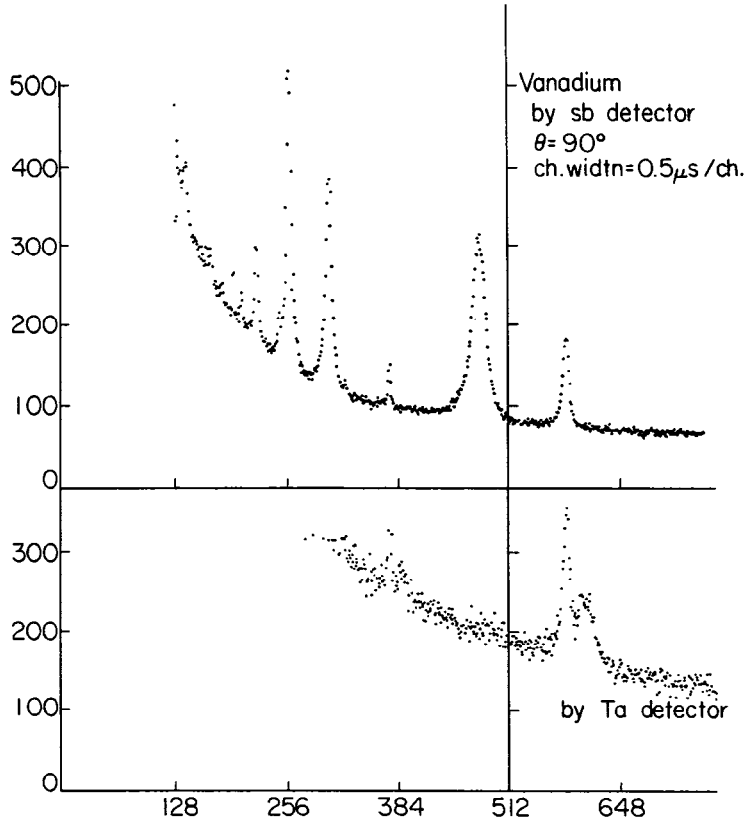


Fig. 5 The scattering from bismuth at 90°, observed at E_f = 6.24 eV with the ^{121}Sb detector. The line is a first-principles calculation, (see text). Dashed line-detector resolution.

first-principles calculation based on the Doppler-broadened scattered neutron profile calculated in Gaussian approximation, on Doppler-broadened and self-shielded resonance capture in the Sb foil, and instrument and source parameters. The agreement indicates that the instrument is well understood.

Tests with vanadium scattering at 90° reveal some potential complications in this type of measurement. Figure 6b shows the result of measurement with the Ta resonance absorber. The two peaks near channel 600 are both due to ^{181}Ta capture - the earlier peak due to capture in Ta, present as impurity in the V sample, the later, broader peak due to capture of scattered neutrons in the resonance absorber foil. By using a ^{121}Sb detector, (E=6.24 eV) we separated the scattered neutron peak from the Ta impurity peak, as shown in Figure 6a. Continuous, 1/v capture in V gives the constant high background in these measurements.



Figs. 6a&b The scattering from vanadium at 90° observed (a) at E_f = 6.24 eV with ^{121}Sb and (b) at E_f = 4.28 eV with ^{181}Ta absorbers.

We measured the scattering from graphite at 90° scattering angle. The data suffered from a substantial background, including the ^{121}Sb peak, presumably because our ^{10}B scintillator and photocathode shield had slipped out of place. The spectrum obtained by removing the Ta absorber proved to be a good measure of the background. Figure 7 shows the net scattering after subtraction of this background, with a first-principles calculation of the scattering to 4.28 eV.

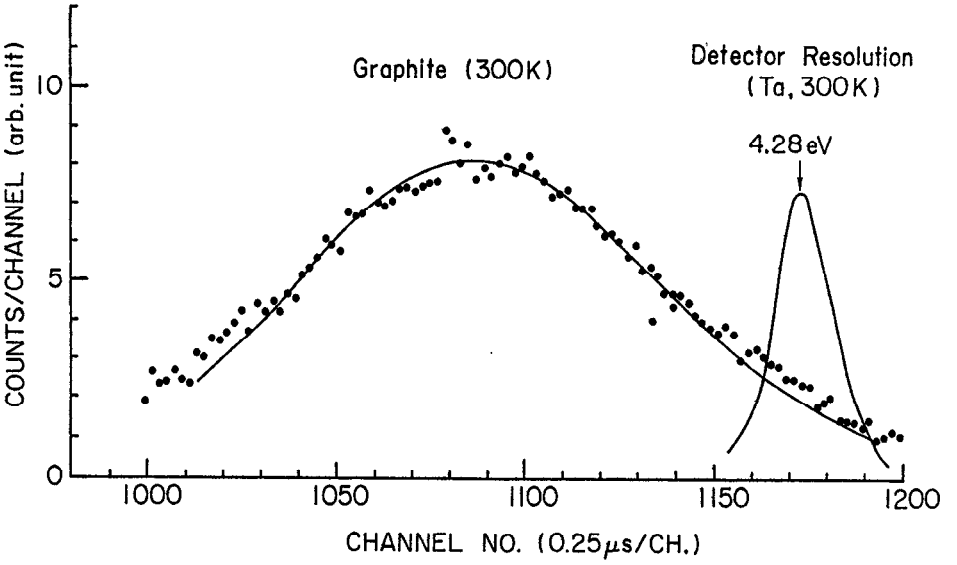


Fig. 7 The scattering from graphite at 90°, observed at E_f = 4.28 eV with ^{181}Ta absorber. The line is a first-principles calculation, modeling the graphite scattering with a gaussian scattering law with an effective temperature of 1097°K.

Satisfactory agreement could only be obtained by calculation using the mean kinetic energy of the struck carbon atoms, near 1100 K, substantially higher than that derived from any of the densities of states that we consulted. The table gives two energy moments of several densities of states which have been presented by different authors. γ_0 is the coefficient of the Debye-Waller factor $e^{-\gamma_0 Q^2}$. T_{eff} is the effective temperature in the Gaussian scattering model $S(Q,E) \alpha \exp[-(\varepsilon-\hbar^2Q^2/2M)^2 / (4\ \hbar^2Q^2/2M)\ k_B T_{eff})]$. Spectra of graphite cannot be successfully predicted using the handbook Debye temperature θ_D=420 K, for which T_{eff} = 325 K. Measurements at 22° scattering angle show a recoilless component, and a low component due to one-phonon scattering. The results have not yet been analyzed.

Table 2 Energy Moments of the Densities of States of Graphite

Density of states	$\gamma_0 eV^{-1}$	$\frac{\bar{E} = 3/2k_B T_{eff}, eV}{T_{eff}, K}$
Young and Koppel[a)]	28.02	$\frac{0.0920}{712}$
Carvalho[b)]	24.70	$\frac{0.0925}{716}$
Page and Haywood[c)]	16.04	$\frac{0.1033}{799}$
Wilson[d)]	29.65	$\frac{0.0977}{756}$
Nicklow Wakabayashi, and Smith[e)]	33.95	$\frac{0.0983}{760}$

a) J. A. Young and J. U. Koppel, J. Chem. Phys. 42, 357 (1965)

b) F. Carvalho, Nucl. Sci. and Eng. 34, 224 (1968)

c) D. I. Page and B. C. Haywood, Atomic Energy Research Establishment (Harwell) Report AERE-R-5778 (1968)

d) J. V. Wilson, Oak Ridge National Laboratory Report ORNL-P-585 (1964)

e) R. Nicklow, N. Wakabayashi and H. G. Smith, Phys. Rev. B 5, 4951 (1972)

We measured the scattering from 300K H_2 gas at 22° scattering angle. Results have not yet been analyzed.

We tested ^{149}Sm and ^{181}Ta absorbers cooled to low temperature, by observing the capture gamma rays produced by the absorber in the sample position. Figure 8 shows the expected narrowing. Both absorbers suffer significant broadening due to self-shielding, and we compute that a 7μm-thick ^{181}Ta foil would provide 70 meV resolution at 50K.

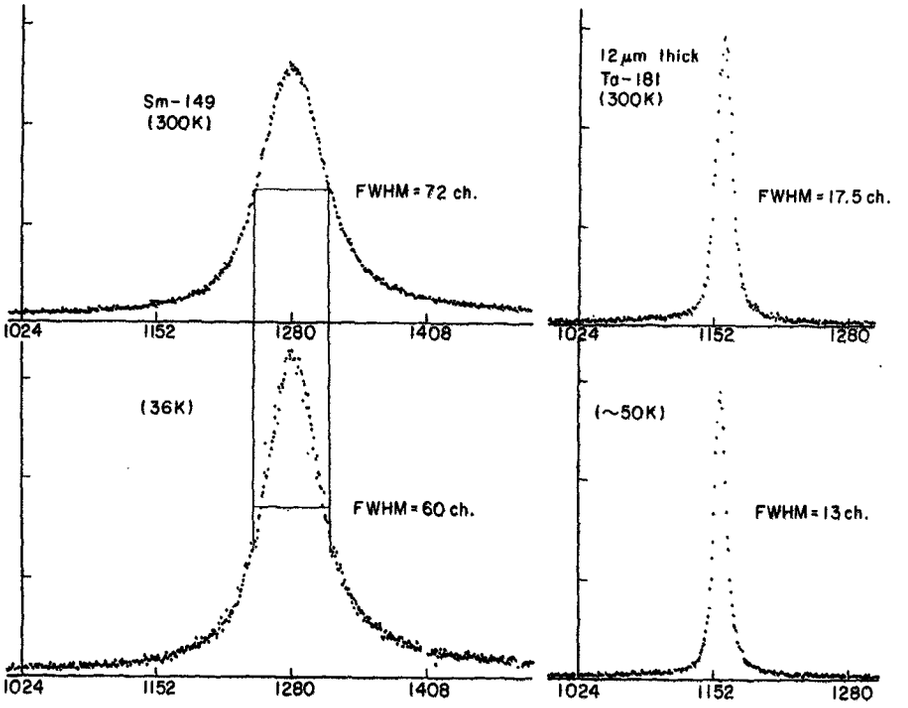


Fig. 8 Measurements with room temperature and cold ^{149}Sm and ^{181}Ta absorbers. On the left are the results for the .87 eV ^{149}Sm resonance (.5 μs/ch), on the right, those for the 4.28 eV ^{181}Ta resonance (.25 μs/ch).

5. CONCLUSIONS

We have solved many of the shielding problems related to resonance detector spectrometers, and measured spectra which are in agreement with calculated expectations. Resolution was in the neighborhood of 100 meV. By use of cooled absorbers of appropriate thickness, resolution can be reduced to about 70 meV, with ^{181}Ta. Further improvements in resolution are possible by use of ^{238}U (E=6.67 eV, Γ=22 meV). Use of a method in which spectra for thick and thin absorbers are subtracted, to eliminate the wings of the resolution function, may provide further improvement.