LA-SUB--97-13

SUMNER ASSOCIATES

100 Cienega, Suite D
Santa Fe, New Mexico 87501
Telephone: 505 984-8041 FAX 505 984-3251

FINAL REPORT
0412J0004-3Y
TASK 44

RECEIVED
JUN 05 1997
OSTI

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

HH
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

Data Reduction for Neutron Scattering from Plutonium Samples

Philip A. Seeger
Sumner Associates
January 31, 1997

Abstract

An experiment performed in August, 1993, on the Low-Q Diffractometer (LQD) at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC) was designed to study the formation and annealing of He bubbles in aged ^{239}Pu metal. Significant complications arise in the reduction of the data because of the very high total neutron cross section of ^{239}Pu , and also because the samples are difficult to make uniform and to characterize. This report gives the details of the data and the data reduction procedures, presents the resulting scattering patterns in terms of macroscopic cross section as a function of momentum transfer, and suggests improvements for future experiments.

1. Introduction

Small-angle neutron scattering may be used to study condensed matter systems on length scales from 1 to 1000 Å. The scattering is sensitive to discontinuities and inhomogeneities in the neutron coherent scattering-length density in the sample. In this experiment, we were looking for information on the size and/or shape of bubbles of He trapped in aged samples of ^{239}Pu . The He is the result of α -decay, so that after 20 years (for instance) we would expect one ^4He nucleus for about every 1740 Pu nuclei in the sample. The coherent scattering length (per atom) for ^{239}Pu is $b = 7.5 \pm 0.3$ fm (or 0.75×10^{-12} cm in the customary units), for a coherent scattering cross section $4\pi b^2 = 7.1 \pm 0.6$ b. The contrast between ^{239}Pu and ^4He ($b = 0.30 \times 10^{-12}$ cm) is high, so scattering from the Pu-He boundaries will be strong. However, the total neutron cross section of ^{239}Pu is 1024.9 ± 2.9 b at a neutron wavelength of 1.798 Å and increases linearly with wavelength. At the longest wavelengths normally used in the LQD the coherent scattering is less than 0.1% of the total cross section. Samples must be thin in order to transmit long-wavelength neutrons. Further, there may be a sample-dependent term in the neutron background due to fission neutrons which are detected asynchronously after moderation in the instrument shielding. These "features" of ^{239}Pu make the experiment and the data reduction challenging.

2. Experiment and Data Acquisition

Three samples were prepared from a source of aged ^{239}Pu (δ -stabilized). One was left at room temperature, the second was annealed at 250 C, and the third was annealed at 400 C. Annealing time was approximately one hour. These are identified respectively as Pu_RT, Pu250,

and Pu400. The thicknesses of the slices measured with a micrometer were 4-1/2 mil, 3 mil, and 6-1/2 mil (or 115 μm , 75 μm , and 165 μm). Since the slices were not smooth, these measurements represent a maximum thickness. The masses of the samples were: Pu_RT, 73.2 mg; Pu250, 59.0 mg; Pu400, (not recorded in LQD lab book). The samples were nominally circular but irregular; the range of diameters should have been 12-15 mm to fit the holders and the beam. Assuming a minimal area of 1.0 cm^2 and a specific volume of 24 $\text{\AA}^3$ per atom, the maximum value for the average thicknesses of Pu_RT and Pu250 would be 44 μm and 36 μm , grossly different from the micrometer measurements. In the future, greater effort should be applied to make the samples uniform, and the area should be measured. These estimated thicknesses will be compared to determinations by neutron absorption below.

The samples were sandwiched in boron-nitride masks and double encapsulated in sample holders with epoxied aluminum windows. An identical empty sample holder was also prepared. The neutron scattering experiments were performed on LQD from August 9-16, 1993. Wavelengths from 0.66-15.3 $\text{\AA}$ were recorded by time of flight in 197 logarithmic slices. The gravity focuser was turned off. The 2-dimensional detector was encoded in 128x128 pixels. Relevant runs are listed in the following table. Because the transmission of the Pu_RT and Pu400 samples was so low at long wavelengths, a second set of transmission runs was made using a mask with twice the transmission of the standard one.

Run #	Archive	Sample	S/T	$\mu\text{A}\cdot\text{h}$	Notes
5248	LQD_930809_103341	none		27.55	Cd-blocked beam
5249	LQD_930809_120029	Pu250	T	101.72	
5254	LQD_930810_090259	Pu250	S	1003.22	
5258	LQD_930816_134047	Pu_RT	S	624.87	LQD_930812_133010 corrected for 2 bad bits
5259	LQD_930812_141416	Pu_RT	T	18.69	
5260	LQD_930812_145152	Blank	T	11.20	
5265	LQD_930813_083202	Blank	S	801.21	
5269	LQD_930813_104143	Pu_RT	T	11.64	big transmission mask
5270	LQD_930813_110001	Blank	T	9.71	big transmission mask
5271	LQD_930813_114532	Pu250	T	11.98	big transmission mask
5272	LQD_930813_122451	Pu400	T	18.01	big transmission mask
5272	LQD_930813_130016	Pu400	T	23.06	
5283	LQD_930815_051948	Pu400	S	1662.04	
5284	LQD_930815_090004	Blank		129.3	Cd-blocked beam
5290	LQD_930816_080652	Pu_RT	S	1104.74	repeat of run 5258

Raw data were extracted from the archived files using the standard LQD data program SMR. The beam center was determined by finding the centroid in the transmission runs, summed over time slices 126–163 (wavelengths 4.86–8.90 Å). For each file, the data in each time-of-flight slice were binned into annular zones one pixel wide centered on the beam position of an appropriate transmission, and a two-dimensional file of counts vs. radius and time (an R - t histogram) was written. For transmission runs ("T" in the S/T column) and for "blocked-beam" runs, a one-dimensional file of counts vs. time of flight was also formed by summing all pixels within 5 pixels of the center in each time slice.

3. Transmissions

Two corrections were made to each transmission run. At early times the instantaneous count rate (especially in the case of the "big" transmission mask) was too high for the detector encoding electronics to keep up. A correction was made assuming that each detected event was followed by a dead time of 20 μ s (chosen to give a smooth response and reasonable ratios). At late times, especially for the samples with highest absorption, count rates become sensitive to room background and so the signal from the blocked-beam run (5284, normalized by μ A-h) was subtracted. The four "big-mask" signals with corrections are shown in Fig. 1. These curves have absorption due to Bragg edges in the approximately 12 mm of Al in the beam path, and also (at early times) individual Bragg lines in the single-crystal MgO filter which is used to attenuate short-wavelength neutrons.

It is also clear that the transmission relative to the blank falls at late times. This is shown in Fig. 2 as ratios to the blank signal. All data for $t > 6000$ μ s are shown and fitted with exponentials. The good linearity is taken as an indication that the livetime and dark-current corrections were done appropriately. The accuracy with which the fits extrapolate to unity at $t = 0$ is a measure of the *a priori* normalization of the data to the μ A-h scaler. The exponential time constant τ shown on Fig. 2 can be related to wavelength, hence to cross section $N\sigma$, hence to effective sample thickness. The (calibrated) total flight path on LQD is 12.72 m. The total cross section for ^{239}Pu at 2200 m/s (or at $t = 5782$ μ s) is 1024.9 ± 2.9 b, hence

$$\sigma(t) = (0.1773 \pm 0.0005 \text{ b}/\mu\text{s}) t$$

$$\text{transmission} = \exp(-N\sigma) = \exp(-t/\tau)$$

$$N = (5.642 \pm 0.016 \mu\text{s}/\text{b}) / \tau$$

where N is the areal density. Again taking the specific volume to be 24 Å^3 ($= 24 \text{ b-cm}$), the ^{239}Pu thickness corresponding to τ is

$$\Delta x = (135.4 \mu\text{s-cm}) / \tau$$

Pu/Bubble Transmission Functions Big mask, with livetime and dark-current corrections

Figure 1. Transmitted-beam signals, corrected for livetime and for dark current, as a function of time of flight. The sum and the fitted line are described in the text.

Sample Transmission Ratios

Figure 2. Ratios of various sample transmissions to the blank. Data from Fig. 1. Fitted exponentials vs. time of flight are used to estimate sample thicknesses.

This estimate of the thickness is an average which is weighted toward the thinner parts of the sample. The big transmission mask sampled the thickness at about eight points on the sample. The resulting values for sample thickness are compared to the previous measurements in the following table.

Sample	Micrometer (μm)	Mass / 1 cm^2 (μm)	$N\sigma(t)$ (μm)
Pu_RT	115	44	64
Pu250	75	36	38
Pu400	165		101

A sum of all the transmission runs with the exponential decays removed (divided out) is shown in Fig. 1. Even with the improved late-time statistics the data are not smooth, so a second-order regression was fitted to all times above 20000 μs . The sum with the fitted tail was then normalized to $\mu\text{A}\cdot\text{h}$ and the exponential decays re-multiplied to give the final transmission functions shown in Fig. 3. It is important to have a set of transmissions with the statistical

Fitted Fluxes (Big Mask)

Figure 3. Smoothed flux functions for the blank and 3 ^{239}Pu samples, normalized per $\mu\text{A}\cdot\text{h}$.

fluctuations between runs removed, as the difference of the reciprocals must be calculated to correct for the blocked-beam contribution to the background.

The usual normalization and background subtraction procedure for LQD data is to divide the recorded histogram for the signal (S) by the transmitted beam (T_S) and subtract the buffer (B) divided by its transmitted beam (T_B), each assumed to be normalized. If however there is a significant difference in the two transmission functions, and if there is also a significant background which is sample independent, then an additional correction term is necessary. Let (K) represent a blocked-beam measurement, and suppose the relative normalization factors ($\mu\text{A}\cdot\text{h}$) of the three measurements are N_S , N_B , and N_K . Then we can subtract the blocked-beam component of the signals before normalizing the transmitted beams as:

$$\begin{aligned} (I) &= \frac{(S) - (K)N_S/N_K}{(T_S)} - \frac{(B) - (K)N_B/N_K}{(T_B)} \\ &= \frac{(S)}{(T_S)} - \frac{(B)}{(T_B)} - \frac{(K)}{N_K} \left[\frac{N_S}{(T_S)} - \frac{N_B}{(T_B)} \right] \end{aligned}$$

The second form is preferred because it allows propagation of errors without correlations, and also shows (K) as a correction to the standard form. Since $(T_S)/N_S$ and $(T_B)/N_B$ are exactly the functions which were fitted in the construction of Fig. 3, the coefficient of (K) can be computed relatively precisely for each of the three samples. The function $1/[N_S/(T_S) - N_B/(T_B)]$ is referred to as a "pseudo-transmission" because it is used to normalize (K) in the same way transmissions are used to normalize other signals.

4. Signals

A transportable Fortran program **QLfromRT** uses the same algorithms (and much of the same coding) as **SMR** to rebin data from an R - t histogram into Q - λ space. The following parameters were used in the transformation:

Flight paths:	$L_1 = 8.40 \text{ m}$, $L_2 = 4.32 \text{ m}$
Detector pixel:	$\Delta R = 3.07 \text{ mm}$, quadratic correction $(1 + 0.0035/\text{pixel})$
Normalization factor:	small mask = 0.0053, big mask = 0.011
Q bins:	linear regime, 20 bins from 0.005 – $0.025 \text{ \AA}^{-1}$ total range 0.0032 – $0.346 \text{ \AA}^{-1}$ (89 bins)
Bin-width criterion:	$\Delta R/R < 1.5 \Delta Q/Q$
Time slices included:	30–188 (3400 – $42000 \text{ }\mu\text{s}$, or $\lambda = 1.06$ – $13.2 \text{ \AA}$)
Radial zones included:	10–70 (28.5 – 270 mm , or $\theta = 0.0066$ – 0.062 rad)

The R - t histograms for the two runs on Pu_RT and for the two blocked-beam runs were added using program **ADD_2D** before conversion to Q - λ . As an illustration of background subtraction procedure, Figs. 4-6 show 2-dimensional Q - λ plots for Pu_RT, the blank, and the blocked beam (normalized with the appropriate pseudo-transmission), including all 197 time (wavelength) slices. Note how similar the blank (Fig. 5) and the signal (Fig. 4) are in the lower right corners, and that the blocked-beam (Fig. 6) has nearly horizontal contours at long wavelengths. The difference is found using program **ADD_2D**, and plotted in Fig. 7. It is seen that for the most part good vertical contours are obtained; *i.e.*, the scattering probability is a function of Q but *not* of λ . However, the earliest and latest time slices are not adequately corrected and have been omitted from the final data. Part of the difficulty appears to be due to the accuracy of determining exposure times (μ A-h), and part may be due to sample-dependent fission-neutron background. Future samples should be made as similar as possible. The scaling of μ A-h at MLNSC has been addressed by the electronics section since this experiment was run in 1993. Better looking subtractions could be obtained by treating the normalization factors as free parameters; for instance, the blank (Fig. 5) could be matched at short wavelengths and the blocked-beam (Fig. 6) at the longest wavelengths. (Such adjustments were *not* made in the plots shown in this report.)

As part of the output of program **QLfromRT**, the Q - λ data are averaged over λ to obtain scattering probability per unit solid angle as a function of Q . Figs. 8-10 show the results for samples Pu_RT, Pu250, and Pu400, respectively, all normalized using the "big mask" transmissions. The blank and blocked-beam were subtracted using program **OPERATE**. The same blank (black dots) is used for all three, but the blocked-beam (cyan dots) has a strong dependence on the sample thickness through the pseudo-transmission function. The Pu400 sample (Fig. 10) was so thick that the blocked-beam correction is excessively large; thus the sample-dependent background determination limits the reliability of the measurement. Error bars shown on all points are standard deviations propagated from counting statistics.

5. Results

For intercomparison of the three samples, the data have been converted to macroscopic cross section (cm^{-1}) by dividing by sample thickness, using the estimates derived from the exponential dependence of transmission with cross section. The results shown in Fig. 11 have also had an estimated "incoherent" contribution of 0.1 cm^{-1} subtracted from each curve. Changing the μ A-h normalization of the Pu400 signal by 3% would eliminate the negatives at high Q without greatly affecting the rest of the curve; I believe this represents the level of systematic error from the beam-monitor scaling. There is considerably more systematic uncertainty in the sample thicknesses, but the data are clearly higher for the Pu250 sample than for either Pu_RT or Pu400.

Figure 4. Scattering probability per unit solid angle from Pu_RT sample, vs. Q and λ .

Figure 5. Scattering probability per unit solid angle from empty holder, vs. Q and λ .

Figure 6. Blocked-beam correction per unit solid angle, vs. Q and λ .

Figure 7. Scattering probability per unit solid angle, Pu_{RT} minus backgrounds.

Figure 8. Signal and background contributions for sample Pu_RT.

Figure 9. Signal and background contributions for sample Pu250.

Figure 10. Signal and background contributions for sample Pu400.

Figure 11. Comparison of 3 samples, converted to macroscopic cross section.

The curve for the unannealed sample, Pu_RT, begins with a slope of -3.0 (on the log-log plot), indicating a very rough interface when viewed on scales greater than $100 \text{ \AA}$. In the mid-range, the slope becomes -3.4 , but then at high Q a new component appears in the scattering, with a slope of -0.9 . If this part of the curve is treated with Guinier analysis, a reasonably good fit (reduced $\chi^2 = 2$) is obtained with a radius of gyration $R_g = 10.6 \pm 0.3 \text{ \AA}$. This suggests a population of bubbles in this size range in sample Pu_RT.

There is no indication that this population of small bubbles remains after annealing at 250 C (sample Pu250). In fact, a power law with a slope of -3.0 fits very well over the entire data range, that is, at all length scales greater than about $10 \text{ \AA}$. This is the form of scattering which would be observed from a very rough surface with a fractal dimension of 3, *i.e.*, a volume filling surface. The higher scattering probability from this sample is also indicative that the He is distributed over a very large surface area.

Sample Pu400 does not display either the population of small bubbles nor the enhanced surface scattering. Within statistical and systematic uncertainties, the scattering looks the same as Pu_RT with the small bubbles removed.

6. Recommendations

Sample uniformity needs to be improved, or at the very least measured. Samples up to the thickness of Pu_RT (whatever it actually is) yield good data. The run times on the samples were adequate to obtain good statistics. More than the usual amount of time should be spent on transmission runs because of the falloff at long wavelengths.

Interesting things seem to have happened in the annealing process; there were gross changes in the scattering. More intermediate temperatures or different durations should be used to try to track the changes in a more linear fashion.

Appendix. Data Files

The names of the data files through the stages of data reduction are as follows.

1. Raw Data

A. Transmissions, from Detector, summed for $R \leq 5$

T_5249.D05
T_5259.D05
T_5260.D05
T_5269.D05
T_5270.D05
T_5271.D05
T_5272.D05
T_5273.D05
T_5284.D05

B. Signals, 2-D histograms vs. R and t

RT5248.DAT
RT5254.DAT
RT5258.DAT
RT5265.DAT
RT5283.DAT
RT5284.DAT
RT5290.DAT
RT5248&84.DAT
RT5258&90.DAT

2. Processed data

LIVEDARK	transmissions with livetime and dark current corrections, as in Fig. 1
FLUXES	fitted fluxes per $\mu\text{A}\cdot\text{h}$, as in Fig. 3
PSEUDOS	pseudo-transmissions
Q_BIG	scattering vs. Q , as in Figs. 8-10
Q_SMALL	scattering vs. Q using small transmission mask
Q_BIG.SMB	signal - blank - blocked beam, as in Figs. 8-10
Q_SMALL.SMB	signal - blank - blocked beam, small transmission mask