

## Measurement of Coherent Neutron Scattering Length with LLL-type Interferometry at PNO in JRR-3M

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Making use of a Si LLL-interferometer with the PNO apparatus at JRR-3M, we successfully determined the coherent neutron scattering lengths of Al, Nb, Ga, W, Cu and Hg, in addition to <sup>69</sup>Ga, <sup>71</sup>Ga, <sup>63</sup>Cu, <sup>65</sup>Cu and <sup>202</sup>Hg, with much higher accuracy than those data in the literature. As for the experimental condition, it was proved that single measurement with wider range of specimen rotation yielded better results than the multiple measurements with narrower range.

**KEYWORDS:** interferometry, coherent scattering length, gallium isotopes, copper isotopes, <sup>202</sup>Hg, W

### §1. Introduction

Neutron scattering lengths of every element and isotope are very important as a fundamental quantity describing the characteristics of the interaction between neutrons and individual nuclei. Their values very widely tabulated in the Koestner-Rauch-Seymann report<sup>1)</sup> are, however, still lacking for several elements and for many isotopes. Around 70 % of all the up-listed values were determined by methods with poor accuracy.<sup>1)</sup> Neutron interferometry is the most powerful tool for this purpose.<sup>2–11)</sup>

In the present article, we are comparing our results obtained with two experimental conditions.<sup>10,11)</sup> For all data in the present article, the most dominant ambiguities will be described, and the base of the determination of the sign for the unknown specimen will be given.

### §2. Experimental Conditions

Neutron experiments were carried out with a Si LLL-type interferometer at the special apparatus "PNO" implemented at JRR-3M.<sup>10,12–14)</sup>

As shown in Fig. 1, every sample was set behind the 1'st reflecting plate (splitter) of the interferometer, receiving both of the transmitted- and the reflected beam, position (a) in Fig.1, or reflected beam only, (b).

The following two experimental conditions were used, the results being compared in the last section;

(1) With the wavelength of 0.15123 nm, the best visibility surveyed being around 44%, specimens were rotated rather smaller angular range, and measurement were repeated near ten times for better statistics.

(2) With the wavelength of 0.15748 nm, we achieved better visibility of around 52%, and specimens were rotated much wider range and measured once.

### §3. Specimens Measured

#### 3.1 Aluminium

As a standard, we measured on aluminium,<sup>10)</sup> which has no isotope. Its purity was 99.99%, and the thickness

was 11.050(0.0005) mm. The atomic weight,  $A$  in eq. (4.4) shown in the next section, of 26.982 g/mol<sup>15)</sup> and the density,  $\rho$  in eq. (4.4), of 2.6993<sup>16)</sup> g/cm<sup>3</sup> were used for the analysis.

#### 3.2 Niobium

We also measured on niobium<sup>10)</sup> as a standard, with no isotope, the purity being 99.95% and the thickness of 2.032(0.0005) mm.  $A=92.91$  g/mol<sup>15)</sup> and  $\rho=8.57$ <sup>16)</sup> g/cm<sup>3</sup> were used.

#### 3.3 Gallium

We measured on the gallium isotopes, because they have only one datum in the literature.<sup>1)</sup> <sup>69</sup>Ga of 99.7% purity and <sup>71</sup>Ga of 99.5% purity were measured.<sup>10)</sup> The isotopes were melted in a warm water bath, and put in flat cells, respectively, made of SiO<sub>2</sub> glass. The resultant thicknesses were 2.000(0.0005) mm for both specimens. In the analysis,  $A=69.72$  g/mol<sup>17)</sup> and

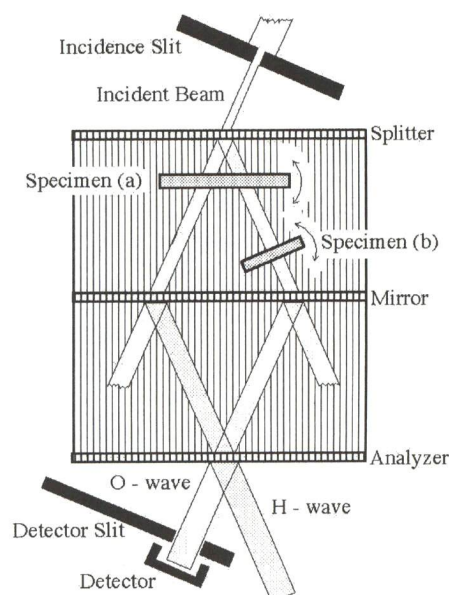


Fig.1. Drawing of the experiment with the beam path.

$\rho=6.0948^{18}$  g/cm<sup>3</sup> for liquid gallium were used.

As a reference, the natural gallium of 99.9999% purity and of 5.000(0.0005) mm thickness was also measured. In this case of solid gallium,  $\rho=5.907^{18}$  g/cm<sup>3</sup> was used.

### 3.4 Tungsten

We measured on a natural tungsten of 99.99% purity,<sup>11)</sup> the thickness being 0.946(0.0005) mm. Its three data in the literature<sup>1)</sup> have large deviation among them.  $A=183.85$  g/mol<sup>15)</sup> and  $\rho=19.3^{19}$  g/cm<sup>3</sup> were used.

### 3.5 Copper

We also measured the copper isotopes, <sup>63</sup>Cu of 99.3% purity and <sup>65</sup>Cu of 99.0% purity, and a natural copper of 99.990% purity,<sup>11)</sup> because only two data are listed up in the literature<sup>1)</sup> for the isotopes with large deviation to each other and large ambiguities. Their thicknesses were 0.543(0.0005), 0.535(0.0005) and 1.924(0.0005) mm for <sup>63</sup>Cu, <sup>65</sup>Cu and the natural copper, respectively. For all cases of copper above,  $A=63.546$  g/mol<sup>15, 17, 19)</sup> and  $\rho=8.96^{15, 19}$  g/cm<sup>3</sup> were used.

### 3.6 Mercury

We also measured one of the mercury isotopes, <sup>202</sup>Hg of 98.80% purity, whose datum is lacking in the literature,<sup>1)</sup> and the natural mercury of 99.9995% purity as a reference.<sup>11)</sup> They were contained within a SiO<sub>2</sub> glass cell of 1.007(0.002) mm thickness, respectively. For all cases of mercury above,  $A=200.59$  g/mol<sup>15, 17, 19)</sup> and  $\rho=13.54616^{15}$  g/cm<sup>3</sup> were used.

## §4. Results and Analysis

### 4.1 Data analysis

Figure 2 are the examples of the interference oscillation curves measured on aluminium(upper), <sup>69</sup>Ga(middle) and <sup>63</sup>Cu (lower), with a least squares fit, respectively. Aluminium and <sup>69</sup>Ga were measured with the experimental condition (1) in the previous section, the former being set at the position (a) in Fig.1 and the latter at (b). <sup>202</sup>Hg was measured with the experimental condition (2), set at the position (b).

Intensity  $I$  of the interference oscillation with the specimen rotation,  $\alpha$ , was analyzed by a least squares method to the following quasi-phenomenological function,<sup>10)</sup>

$$I = A \cos\left\{ \frac{B}{\cos(\theta_B + \alpha + C)} - \frac{B}{\cos(\theta_B - \alpha - C)} + D \right\} + E, \quad (4.1a)$$

$$I = A \times \cos\left\{ \frac{B}{\cos(\alpha + C)} + D \right\} + E, \quad (4.1b)$$

the former being for the both-beam case, (a) in Fig.1, the latter for the one-beam case of (b). Among the five fitting parameters, A - E in equations above, only  $B$  includes all the physical terms as:

$$B = -N \times b \times \lambda \times t, \quad (4.2)$$

where  $N$  : atomic number density of the sample,  $\lambda$  : wavelength and  $b$  : the coherent scattering length to be determined and  $t$  : the thickness of the specimen.

When we take account of the effect of the air displaced

by the sample, eq. (4.2) is modified as follows,

$$B_{\text{sample}} = B_{\text{expr}} + B_{\text{air}}. \quad (4.3)$$

Here, the  $N$  was estimated practically<sup>10)</sup> by

$$N = N_A \times \rho / A, \quad (4.4)$$

with  $N_A$ : the Avagadro's constant,  $\rho$  : the density and  $A$ : the atomic weight for the specimen, respectively.

The primary value of the coherent scattering length,  $b$ , is thus derived as following relation;

$$b = B_{\text{sample}} / (N \times \lambda \times t), \\ = B_{\text{expr}} / (N \times \lambda \times t) + b_{\text{air}} \times N_{\text{air}} / N_{\text{sample}}. \quad (4.5)$$

With the atomic constitutions<sup>18)</sup> of 0.781 of nitrogen,  $b_N=6.44$  fm,<sup>1)</sup> 0.209 of oxygen  $b_O=5.83$  fm,<sup>1)</sup> and 0.009 of argon,  $b_A=2.07$  fm,<sup>1)</sup> respectively,  $b_{\text{air}}$  was estimated as 16.907(0.033) fm, and  $N_{\text{air}} = 2.69 \times 10^{19}$  atoms/cm<sup>3</sup> at T=295 K with the gas-constant of  $R=82.0568$  cm<sup>3</sup>×atm/(mol)/K.<sup>18)</sup>

The primary  $b$ -value, thus obtained, was refined first by removing the effect of impurities. It was refined further and finally by eliminating the effect of the isotopic mutual contamination, if concerned.

Ambiguities for final  $b$ -values were estimated using eq. (4.5) in company with eq. (4.4);

$$\frac{\delta b}{b} = \frac{\delta B_{\text{expr}}}{B_{\text{expr}}} + \left( \frac{\delta N_A}{N_A} + \frac{\delta \rho}{\rho} + \frac{\delta A}{A} \right) + \frac{\delta \lambda}{\lambda} + \frac{\delta t}{t}, \quad (4.6)$$

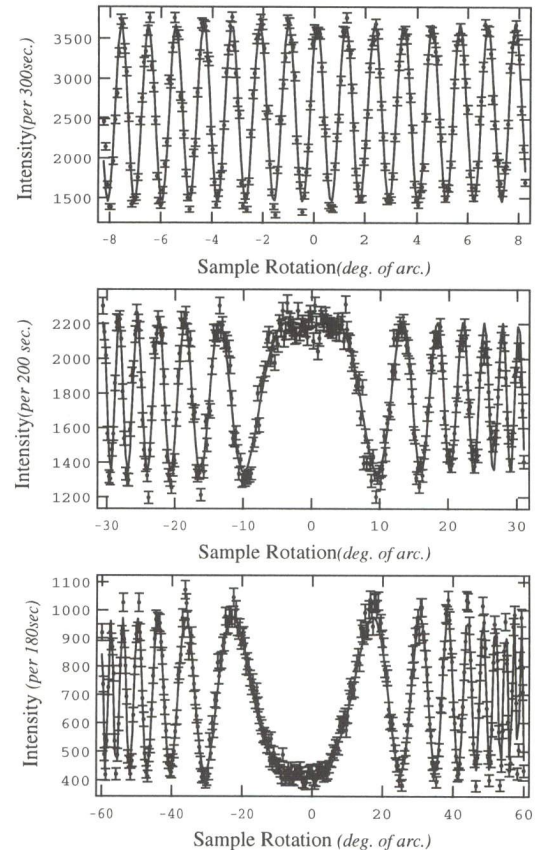


Fig.2. Example of the interference oscillation curves of aluminium (upper), <sup>69</sup>Ga(middle) and <sup>63</sup>Cu (lower), with a least squares fit, respectively.

where the contribution of air was less than  $10^{-5}$  and neglected. The most dominant factor is shown for every specimen in Table I, where we assumed that the data given without any ambiguity contained the measuring fractions of .5 in the place of the last decimal.

#### 4.2 Sign of $b$

We should be careful determining the sign of  $b$  of an unknown sample. In the case of plural specimens of the same sign of  $B_i$ 's, the total phase term  $B$  in the eq. (4.1a) or eq. (4.1b) is generally expressed as follows;

$$B_{obs} = |B| = |\Sigma B_i| = \Sigma |B_i|, \quad (4.7a)$$

while for mixed signs, because of more or less cancellation among them;

$$B_{obs} = |\Sigma B_i| < \Sigma |B_i|. \quad (4.7b)$$

In our case for  $^{202}\text{Hg}$ , we measured three  $B_{i,obs}$ 's below and confirmed the relations among them;

$$B_{(sample+cell)+(cell),obs} = B_{(sample+cell),obs} + B_{(cell),obs}, \quad (4.8a)$$

$$B_{(sample+cell),obs} > B_{(cell),obs}. \quad (4.8b)$$

eq. (4.8a) obviously corresponds to eq. (4.7a), *i.e.* the sign of  $B_{(sample+cell)}$  is the same to that of  $B_{(cell)}$ , which has the following two possibilities;

- (1)  $B_{(sample)}$  has the same sign with  $B_{(cell)}$ ,
- (2)  $B_{(sample)}$  has the opposite sign to  $B_{(cell)}$  but  $|B_{(sample)}|$  is smaller than  $|B_{(cell)}|$ , then obviously;

$$B_{(sample+cell),obs} = |B_{(sample)} + B_{(cell)}| \\ = |B_{(cell)}| - |B_{(sample)}| < |B_{(cell)}| = B_{(cell),obs}. \quad (4.9)$$

The latter "possibility" (2) is, however, denied immediately, because eq. (4.9) obviously conflicts with one of the observed facts, eq. (4.8b). Consequently, the sign of  $b_{(sample)}$  was concluded to be the same to that of  $b_{(cell)}$ .<sup>11)</sup>

### §5. Conclusion and Summary

Making use of a Si LLL-type neutron interferometer on the PNO-apparatus at JRR-3M, we successfully determined the coherent neutron scattering lengths of aluminum, niobium, gallium, tungsten, copper and mercury, in addition to their isotopes of  $^{69}\text{Ga}$ ,  $^{71}\text{Ga}$ ,  $^{63}\text{Cu}$ ,  $^{65}\text{Cu}$  and  $^{202}\text{Hg}$ . The results obtained are summarized up in Table I compared with the literature.<sup>1)</sup> In the Table I, a brief expression (2+b), for example, means that the sample was measured with the experimental condition (2) and with the reflected beam only.

Through the present neutron-interferometric measurements, it can be concluded that;

(1) The  $b$ -values were determined with higher accuracy than those in literature,<sup>1)</sup> and improved the accuracy by one or two figures.<sup>10,11)</sup>

(2) Specimens could be set in both-beam, position (a) in Fig.(1), or in single-beam, (b), depending on the specimen size.

(3) The sign of  $b$  of an unknown sample was easily determined by a simple mathematical consideration.<sup>11)</sup>

Table I. Summary of the coherent scattering lengths measured,  $b$  in fm, comparing with the tabulated values,<sup>1)</sup> together with the experimental conditions and the most dominant factor against the ambiguity. See text.

| Specimen          | condition | b(measured)    | ambiguity                | b(Table)     |
|-------------------|-----------|----------------|--------------------------|--------------|
| Al                | (1+a)     | 3.427(0.004)   | $\delta B_S/B_S=0.0013$  | 3.44-3.445   |
| Nb                | (1+a)     | 7.128(0.011)   | $\delta B_S/B_S=0.0016$  | 6.9-7.14     |
| nat.Ga            | (1+a)     | 7.212(0.014)   | $\delta B_S/B_S=0.0019$  | 7.2-7.288    |
| $^{69}\text{Ga}$  | (1+b)     | 8.053(0.013)   | $\delta B_S/B_S=0.0029$  | 7.88         |
| $^{71}\text{Ga}$  | (1+b)     | 6.170(0.011)   | $\delta B_S/B_S=0.0035$  | 6.40         |
| nat. W            | (2+b)     | 4.7555(0.0181) | $\delta\rho/\rho=0.0026$ | 4.77-5.1     |
| nat.Cu            | (2+b)     | 7.7093(0.0086) | $\delta\rho/\rho=0.0006$ | 7.35-7.90    |
| $^{63}\text{Cu}$  | (2+b)     | 6.477(0.013)   | $\delta t/t=0.0009$      | 6.40, 6.72   |
| $^{65}\text{Cu}$  | (2+b)     | 10.204(0.020)  | $\delta t/t=0.0009$      | 10.57, 11.09 |
| nat.Hg            | (2+b)     | 12.595(0.045)  | $\delta t/t=0.0020$      | 12.66-13.10  |
| $^{202}\text{Hg}$ | (2+b)     | 11.002(0.043)  | $\delta t/t=0.0020$      | none         |

(4) Ambiguities of the derived  $b$ 's with narrow angular rotation of specimen were dominantly restricted by the experimental error itself,  $\delta B_S$ .

(5) Ambiguities with wider rotation of specimen, on the other hand, were dominantly restricted by the accuracies of the "table values", such as the density and/or the atomic weight, in addition to the specimen thickness, because the experimental error itself was rather small.

(6) In the sense of ambiguity in  $B$ -value, consequently, measurement with enough angular range of specimen rotation is more desirable, even if it is only once-measurement, than the multiple measurements with narrower range and with longer period of measurement.

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