# Neutron Coherent Scattering Amplitudes for Cd and Eu†

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The neutron coherent scattering amplitudes of Cd and Eu have been measured from diffraction studies on CdO and $Eu_2O_3$ at a neutron wavelength of 1.391 A. Using the value of $0.58\times10^{-12}$ cm as the scattering amplitude for oxygen, the experimental values for the Cd and Eu coherent scattering lenths are $|f_{Cd}| = (0.58 \pm 0.03)\times10^{-12}$ and $|f_{Eu}| = (0.55\pm0.04)\times10^{-12}$ cm. The experimental values for the nonresonant coherent scattering lengths are $f_0(Cd) = (0.78\pm0.04)\times10^{-12}$ cm and $f_0(Eu) = (0.80\pm0.05)\times10^{-12}$ cm. The phase of the scattering for both elements is positive.

## INTRODUCTION

VALUES of coherent scattering amplitudes for elements which possess high thermal neutron absorption cross sections have been determined in relatively few cases.[1] The diffracted neutron intensity obtained from these elements is minimized on account of the high absorption in the sample and the small diffracted intensity arising from the thin samples which are dictated by the large absorption. Therefore, diffraction data having the necessary statistical accuracy for determining scattered amplitudes are accumulated slowly for these elements. The data are usually characterized by a low peak-to-background intensity ratio and in the observance of selected planes having relatively high multiplicity values.

Utilizing one of the neutron diffractometers at the Omega West Reactor, it has proved feasible to obtain diffraction data from these highly absorbing elements by using the sample in transmission and employing special types of sample holders. The spectrometer is conventional and uses the (111) planes of a single lead crystal to produce a flux of about $10^7$ neutrons/cm$^2$-sec at a wavelength of 1.391 A. Collimators for the incident and diffracted beams give a half-maximum width of about one degree for a peak at a 40° scattering angle. The beam incident on the sample is monitored by a transmission fission counter. A $BF_3$ counter is employed as a neutron detector. Oxides of the elements have proved most satisfactory for measuring scattering amplitudes. The metals themselves have yielded inconsistent results; the cause of this is probably due to preferential orientation of crystal planes produced in rolling or pressing the necessary thin foils.

## EXPERIMENTAL

Two independent sets of measurements were made with CdO to obtain data for the Cd scattering amplitude. One measurement was taken with the CdO powder (about 0.1 g/cm$^2$) pressed on a $\frac{1}{32}$ in. thick Ti-Zr alloy disk as a holder. A background run observed with the Ti-Zr holder alone showed no diffraction peaks. The other measurement was made using a disk which had been pressed from a mixture of CdO powder and vanadium metal powder; the disk consisted of about 25% by weight of CdO (0.06 g/cm$^2$). A background run made with only the vanadium metal exhibited only one diffraction peak at 38°.

The oxide, $Eu_2O_3$, was used in the Eu measurement and had a purity of 99%. As in the Cd case, the oxide (0.11 g/cm$^2$) was mixed 25% by weight with vanadium metal powder and the resulting mixture pressed into a disk. The neutron transmission through this disk, as well as in the CdO case, was approximately 40%.

Nickel powder, for which the scattering amplitude is $1.05\times10^{-12}$ cm, was used as a standard in evaluating the geometrical constant $K$ in the expression for the diffraction peak intensities,[2]

$$P_{hkl}=K\rho' t \exp(-\mu t \sec\theta) j_{hkl} N_c^2 F_{hkl}^2/\rho \sin^2 2\theta. \quad (1)$$

In the beginning of these experiments, this constant was independently verified by using $ThO_2$ as another standard. However, during the period of taking data, the nickel standard was occasionally substituted for the oxide samples and the constant evaluated. The lattice parameter, $a=4.695$ A, was used for CdO and $a=10.864$ A was used for $Eu_2O_3$.

## RESULTS AND DISCUSSION

### Cd

A typical set of diffraction data obtained from the CdO supported by the Ti-Zr alloy holder is illustrated in Fig. 1. Only three diffraction peaks were observed from this oxide but all are well resolved and usable. The peaks are superimposed on a high background with the maximum peak height being 75% above background level. A similar diffraction pattern was obtained from the CdO supported in vanadium metal except for the form of the base line.

The structure factor of CdO for the diffraction peaks observed here (even values of *hkl*) is given by

$$F_{hkl}=4(f_{Cd}+f_O)\exp[-W(T)\sin^2\theta_{hkl}/\lambda^2], \quad (2)$$

† This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] Previous work relating to the coherent scattering length for Cd has been done by S. W. Peterson and H. G. Smith, Phys. Rev. Letters **6**, 7 (1961) and by B. N. Brockhouse and A. T. Stewart (unpublished).

[2] G. E. Bacon, *Neutron Diffraction* (Oxford University Press, New York, 1955).

in which the nomenclature is standard.[2] The planes corresponding to odd values of *hkl* (e.g., 111 and 311) contain the term $f_{Cd}-f_O$ in their structure factor. The customary method[3] was used for evaluating $F$ at $\theta=0°$ and is illustrated in Fig. 2 where $F^2$, as calculated from Eq. (1), is plotted on a log scale against $\sin^2\theta/\lambda^2$. The straight line in Fig. 2 has been drawn through a weighted average of the two sets of measurements from CdO. The extrapolation of this line to $\theta=0°$ gives an intercept at 0° of $F^2=(2.15\pm0.1)\times10^{-23}$ cm$^2$. If $f_{Cd}$ is calculated from this intercept and using Eq. (2), one obtains $f_{Cd}=(0.58\pm0.03)\times10^{12}$ cm assuming that $f_O=0.58\times10^{-12}$ cm.

The above simplified procedure for calculating the scattering amplitude is not correct if the neutron energy is within the region of a resonance where resonance and potential scattering interfere with each other. Following the procedure used in the case of x rays near an absorption edge,[4] the scattering length for neutrons may also be expressed in complex form as $f=f_0+\Delta f'+i\Delta f''$, where $f_0$ is the nonresonant coherent scattering length, $\Delta f'$ is the real resonant increment, and $\Delta f''$ the imaginary resonant increment. The amplitude $f_0$ is that which should be observed at wavelengths far removed from the resonance region and which is associated with the potential scattering $\sigma_p=4\pi f_0^2$. Peterson and Smith[1] have recently listed the explicit expressions for $\Delta f'$ and $\Delta f''$ in the case of neutrons. In the present experiment, the resonance at 0.18 ev from the $Cd^{113}$ isotope makes it necessary to express the Cd scattering length in the above complex form. Applying the expressions and parameters listed for $\Delta f'$ and[1] $\Delta f''$ at $\lambda=1.391$ A, one obtains $\Delta f'=-0.20\times10^{-12}$ cm and $\Delta f''=-0.08\times10^{-12}$ cm. Equation (2) then gives $f_0(\mathrm{Cd})=(0.78\pm0.04)\times10^{-12}$ cm. The present measurement is higher than the value of $f_0(\mathrm{Cd})=(0.62\pm0.03)\times10^{-12}$ cm measured by Brockhouse and Stewart.[5] The cadmium nuclear radius, $R_{Cd}$, should be approximately

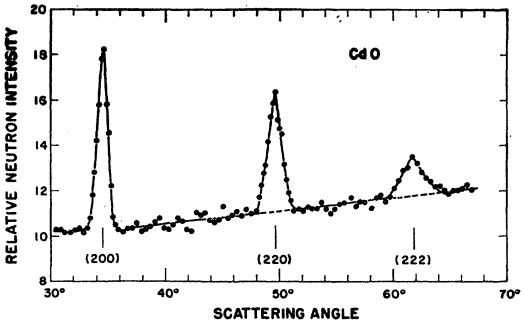


FIG. 1. Neutron diffraction results from CdO contained in Ti-Zr alloy holder.

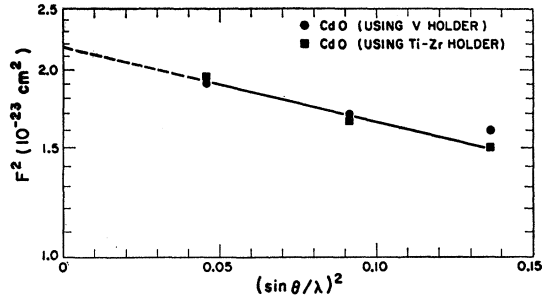


FIG. 2. Graphical method for evaluating $F^2$ at zero scattering angle. $F$ is the crystal structure scattering amplitude.

equal to $f_0(\mathrm{Cd})$. Using the expression $R=1.5\times10^{-13}A^{\frac{1}{3}}$, the result is $R_{Cd}=0.72\times10^{-12}$ cm which lies between the above two experimental values for $f_0(\mathrm{Cd})$. The absolute value of the coherent scattering amplitude derived from the complex form, $f_{Cd}=(0.78-0.20-i0.08)\times10^{-12}$ cm, is $|f_{Cd}|=0.585\times10^{-12}$ cm. Due to the small value of the imaginary component of $f_{Cd}$, the latter result is very close to the value obtained from the approximate method used in the previous paragraph. Values for $f_{Cd}$ or $|f_{Cd}|$ will, of course, depend upon the incident neutron wavelength over the neutron energy range where the increments $\Delta f'$ and $\Delta f''$ make appreciable contributions to the coherent scattering length.

The phase of the coherent scattering from Cd is obviously positive as judged from the fact that peaks corresponding to odd values of *hkl* are not observed (or are extremely weak) in the diffraction pattern.

## Eu

The diffraction pattern obtained from $Eu_2O_3$ supported in vanadium is shown in Fig. 3. Each data point presents the sum of several experimental runs and has a statistical accuracy of 1% or better. This oxide shows several diffraction peaks with the maximum peak height being 50% above background level; the rise of the base level at the left of the pattern is due to instrumental conditions. Only the peaks originating from the (222) and (332) planes are resolved in these data; the other peaks are composed of two or more closely spaced reflections.

The structure factor for $Eu_2O_3$, which was computed by means of standard formulas,[6] is a function of the three values of the oxygen position parameters $(x,y,z)$ and the europium position parameter, $u$. These four parameters have not been directly measured for the present oxide. However, values obtained in conjunction with a neutron diffraction study[7] of $Ho_2O_3$ and direct measurements of the position parameters of the mineral bixbyite,[8,9] $(Fe, Mn)_2O_3$, are probably close to values

[3] C. G. Shull and E. O. Wollan, Phys. Rev. **81**, 527 (1951).
[4] See, e.g., R. W. Jones, *The Optical Principles of the Diffraction of X-Rays* (G. Bell and Sons, London, 1948), p. 149.
[5] According to a private communication, this value was obtained by B. N. Brockhouse and A. T. Stewart in 1954 using CdO powder in reflection at a neutron wavelength of 1.17 A.
[6] *International Tables for X-Ray Crystallography* (Kynock Press, Birmingham, 1952), Vol. I.
[7] E. V. Koehler, E. O. Wollan, and M. K. Wilkinson, Phys. Rev. **110**, 37 (1958).
[8] H. Dachs, Z. Krist. **107**, 370 (1956).
[9] L. Pauling and M. D. Shappell, Z. Krist. **75**, 128 (1930).

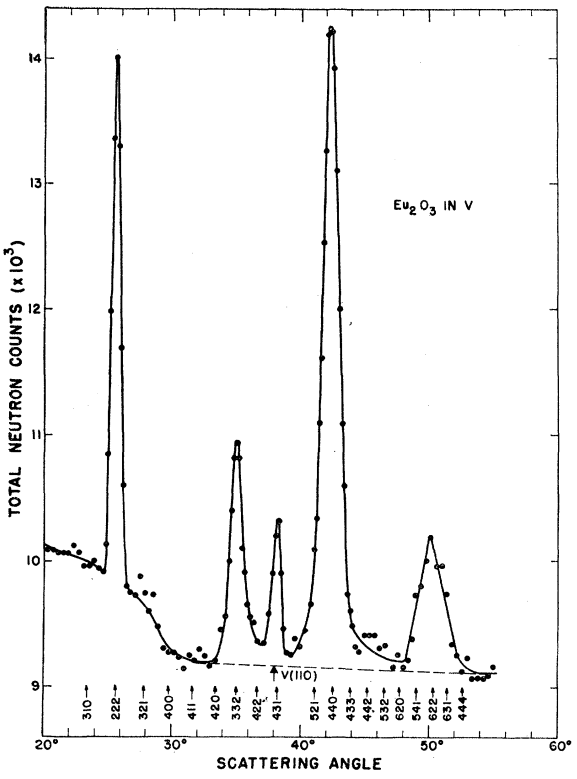


FIG. 3. Neutron diffraction pattern obtained from $Eu_2O_3$ supported in vanadium metal powder.

associated with $Eu_2O_3$. Fortunately, the intensity from the (222) plane is insensitive to the oxygen parameters and only weakly dependent on the europium parameter. Furthermore, the diffraction data in Fig. 3 show that the peak from the (222) plane is best resolved and that the base line can be accurately determined for this peak. For the above reasons, the (222) plane has been given the most weight in the evaluation of the Eu scattering amplitude.

Since Eu has a number of resonances in the region of the neutron wavelength, the complex notation must be used for the Eu scattering length. Table I lists calculated values of $\Delta f'$ and $\Delta f''$ for the resonances which

TABLE I. Calculated values of $\Delta f'$ and $\Delta f''$ for Eu resonances.

| Isotope | $E_0$(ev) | Calc. $\Delta f'$ ($10^{-12}$ cm) | Calc. $\Delta f''$ ($10^{-12}$ cm) |
|---|---|---|---|
| $Eu^{151}$ | 0.327 | −0.029 | −0.0036 |
| $Eu^{151}$ | 0.461 | −0.150 | −0.0166 |
| $Eu^{151}$ | 1.055 | −0.014 | −0.0001 |
| $Eu^{151}$ | 2.71 | −0.0060 | −0.003 |
| $Eu^{151}$ | 3.37 | −0.0030 | −0.00005 |
| $Eu^{151}$ | 3.71 | −0.0010 | −0.00002 |
| $Eu^{153}$ | 1.76 | −0.0030 | −0.0001 |
| $Eu^{153}$ | 2.46 | −0.039 | −0.0007 |
| $Eu^{153}$ | 3.29 | −0.0019 | −0.00003 |
| $Eu^{153}$ | 3.94 | −0.0016 | −0.00002 |
| | | $\Sigma\Delta f' = -0.248$ | $\Sigma\Delta f'' = -0.024$ |

contribute significantly to these increments; summing these values gives the result $f_{Eu} = f_0(Eu) - 0.25 - i0.02$. A spin weighting factor of $g = \frac{1}{2}$ was used in these calculations; other required parameters were obtained from a standard reference.[10]

Table II gives structure factor expressions for various $Eu_2O_3$ reflecting planes and also the experimental values for $F^2$. Using these data in Eq. (1) gives the results for $f_0(Eu)$ listed in Table II. The value, $f_0(Eu) = 0.80 \pm 0.05$ as derived from the (222) plane, is considered the most reliable. Lower values for $f_0(Eu)$ are obtained from the (332) and (440) planes. These lower figures appear logical since the base level is uncertain for these planes and is probably drawn too high. The $f_0(Eu)$ value derived from the (332) peak is very sensitive to the base line level; in the case of the (440) peak, the result is not as sensitive to the error in the base line level but an additional error is present from estimating the correction to the (440) area from the contribution of the nearby (521) peak. No attempt was made to utilize the diffraction peak occurring at 50° on account of the many unresolved reflections. The experimental result for $f_0(Eu)$ agrees fortuitously well with the Eu nuclear radius calculated as $R_{Eu} = 1.5 \times 10^{-13} A^{\frac{1}{3}} = 0.80 \times 10^{-12}$ cm. The absolute value of the Eu coherent scattering length, $|f_{Eu}|$, at $\lambda = 1.391$ A is $(0.55 \pm 0.04) \times 10^{-12}$ cm. No temperature correction has been applied to the above Eu scattering amplitudes. However, this correction was estimated to be small and within the experimental error on account of the small angle at which the (222) reflection occurs.

The above results are based on structure factors computed from position parameters of $x = 0.370$, $y = 0.155$, $z = 0.405$, and $u = -0.030$. Interatomic europium to oxygen separation distances calculated from these position parameters lead to a minimum value of 2.14 A and a maximum value of 2.60 A; the minimum value is a reasonable distance, but the maximum value appears slightly high. The above set of position parameters were found to give the most consistent set of values for $f_0(Eu)$ derived from the different planes. However, as previously mentioned, using other accepted sets of position parameters changes the value of $f_0(Eu)$ very little in the case of the (222) plane. For example, the $Ho_2O_3$ parameters[7] ($x = 0.395$, $y = 0.150$, $z = 0.385$, $u = -0.030$) give $f_0(Eu) = 0.78 \times 10^{-12}$ cm and the bixbyite parameters[9] ($x = 0.385$, $y = 0.145$, $z = 0.380$, $u = -0.030$) give $f_0(Eu) = 0.79 \times 10^{-12}$ cm. Using either of the latter two sets of position parameters to calculate $f_0(Eu)$ from the (332) and (440) planes resulted in considerably lower values than those listed in Table II.

The phase of the Eu scattering can be established from the presence or absence of diffraction peaks from certain reflecting planes. For example, no peak is ob-

[10] *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.

TABLE II. Computed values of $f_0$(Eu) derived from different diffraction peaks.

| $hkl$ | Scattering angle | Structure factor[a] | Multiplicity | $P_{hkl}$[b] | $F_{hkl}^2$ ($10^{-12}$ cm$^2$) | $f_0$(Eu) ($10^{-12}$ cm) |
|---|---|---|---|---|---|---|
| 222 | 25.6° | $0.43\ f_0-30.31\ f_{Eu}$ | 8 | 593± 20 | 274± 30 | 0.80±0.05 |
| 332 | 35.0° | $14.90\ f_0+\ 2.95\ f_{Eu}$ | 24 | 366± 25 | 101± 20 | 0.72±0.08 |
| 440 | 42.3° | $31.60\ f_0+27.67\ f_{Eu}$ | 12 | 1320±100[c] | 1020±200 | 0.74±0 08 |

[a] Structure factor calculated for $x=0.370$, $y=0.155$, $z=0.405$, and $u=-0.030$. The Debye-Waller temperature factor has been omitted in this tabulation.
[b] $P_{hkl}$ is proportional to the area under the diffraction peaks.
[c] This value of $P_{440}$ has been corrected for the contribution of the adjacent (521) peak.

served at the Bragg angle corresponding to the (400) plane; since the structure factor is $F_{400}\cong 30f_{Eu}-45f_O$, it can be concluded that Eu scatters with the same phase as oxygen or positive phase. Additional confirmation of the phase is established by the large peak at 42.3° which is primarily due to the (440) plane; the structure factor, $F_{440}=27.7f_{Eu}+31.6f_O$, again indicates that Eu and O scatter with the same phase.


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# Single-Quantum Annihilation of Positrons*


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An experiment has been performed to investigate single-quantum annihilation of positrons in the field of high-$Z$ nuclei. This mode of annihilation has been observed and the absolute magnitude, energy dependence, and $Z$ dependence of the cross section measured. Backgrounds which might contaminate the derived signals have been semiempirically investigated and found to be an order of magnitude or more below the rate identified as single-quantum annihilation. The data on absolute magnitude of the counting rate and its dependence on positron energy agree with the calculations of Jaeger and Hulme. The $Z$ dependence agrees with a cross section proportional to $Z^5$.


## I. INTRODUCTION

ANNIHILATION of an electron-positron pair can take place via the emission of one, two, three, or more quanta. In solids, the most probable mode is two-quantum annihilation of a positron electron pair almost at rest.[1,2] This accounts for all but a few percent (e.g., 3%) of positrons from typical radioactive sources, when the positrons are stopped in metals. In this case, two-quantum annihilation in flight[1,3,4] accounts for most of the remaining positrons.

Three-quantum annihilation occurs from the $^3S_1$ state of the $e^+e^-$ system. This is a consequence of conservation of angular momentum, and the transverse nature of the electromagnetic field. Three quantum annihilation is profuse in those gases (e.g., argon) not capable of causing transitions to the more rapidly annihilating $^1S_0$ state. Indeed, under certain conditions of external field[5] annihilation via three quantum emission occurs nearly 75% of the time.

There are no other profuse annihilation modes. Annihilation via the emission of more than three quanta

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[5] M. Deutsch, *Progress in Nuclear Physics* (Butterworths Scientific Publications, Ltd., London, 1953), Vol. 3.