TABLE IV. $E2/M1$ mixing ratio for the 23.875-keV transition from L subshell ratios.

Subshell ratio	Measured ratio	$\delta^2=E2/M1$ [a] Rose	 Sliv and Band	 Hager and Seltzer
L_1/L_2	12.35 ± 0.3	$(2.05\pm0.72)\times10^{-4}$	$(\leq1.75)\times10^{-4}$	$\leq1.19\times10^{-4}$
L_1/L_3	$45.7\ \pm1.5$	$(0.82\pm0.13)\times10^{-4}$	$(\leq0.36)\times10^{-4}$	$\leq0.33\times10^{-4}$

[a] In the calculation of these values, account was taken of the uncertainties in the experimental intensity ratios and of an assumed uncertainty in the theoretical ratios for order $M1$. The latter was taken to be 3%, except in the case of L_1:L_3 from Sliv and Band (Ref. 10), where 5% was used, for a reason given in Sec. IV B.

Relativistic corrections for these nonrelativistic L_1, M_1, N_1, and O wave functions are all very nearly the same.

D. Total Conversion Coefficient of the 23.875-keV Transition

Because of the difference in line shape between $K65.66$ and the other lines, it was not possible accurately to sum line intensities of the 65.66-keV transition and compare this sum with the intensity sum from the 23.875-keV transition in order to obtain α_{total} (23.875). One can, however, obtain the α_{total} value from the sum of the observed 23.875-keV intensities (Table III) by assuming the theoretical value for any one line. Thus, if one takes L_1 23.875 to be 3.75 ± 0.07, α_{total} (23.875) is found to be 5.12 ± 0.14. In this value the O intensity is included; M_4 and M_5 make negligible contributions. One may compare this value for α_{total} with that of Benczer-Koller[3] 5.2 ± 0.3.

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Spin Dependence of the U^{235} Low-Energy Neutron Cross Section*

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The interaction of polarized monochromatic neutrons with polarized U^{235} nuclei has been used to study the spin dependence of the total cross section of U^{235} at a number of neutron energies between 0.075 and 2.04 eV. Measurements were made primarily with a $^{235}U_{0.2}La_{0.8}Cl_3$ target in which appreciable nuclear polarization was produced. The results show that the 0.275-eV resonance is in the opposite spin state to the 1.14-eV resonance and to the major part of the thermal cross section. The 2.08-eV resonance is probably in the same spin state as the 0.275-eV resonance. If we assume that U^{235} has a negative magnetic moment, then $J=I-\frac{1}{2}=3$ at 0.275 eV. None of the existing cross-section analyses are completely consistent with the observations for $E\leq1.14$ eV. In U^{235} metal a small "brute-force" nuclear polarization was detected, but there was no evidence of any hyperfine interaction. A somewhat larger nuclear polarization was observed in $^{235}UFe_2$, due to a negative hyperfine field of undetermined magnitude.

I. INTRODUCTION

ALTHOUGH the low-energy neutron cross section of U^{235} has been the subject of intensive study, no completely satisfactory analysis of it in terms of resonance theory has been possible. The difficulty originates in two basic facts: first, fission is a few channel process; second, the fission widths of the levels are comparable to their spacings. Thus there is appreciable interference between resonances in the same spin state and as a consequence the simple Breit-Wigner single-level formula is inadequate for the resonance analysis.[1] This is a situation in which a more general approach such as the Wigner-Eisenbud formalism[2] must be used. In principle, the resulting multilevel analysis should provide the basis for a complete description of the cross section; however, in practice, there is no systematic way to apply it to U^{235}. The problem is that the levels which interfere have to be specified in order to get a unique set of parameters from the analysis. Unfortunately, there is no objective procedure, based on the cross-section data alone, for grouping the resonances according to spin.

The large thermal cross section of U^{235} is also difficult to explain. It is usually attributed to one or more compound states lying just below the neutron binding energy, i.e., "negative energy" resonances. However, if only

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[1] V. L. Sailor, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 4, p. 199.

[2] E. P. Wigner, Phys. Rev. **70**, 606 (1946); E. P. Wigner and L. Eisenbud, *ibid.* **72**, 29 (1947).

one such state is responsible, its strength must be abnormally large to account for the magnitude and energy dependence of the observed thermal cross section. Of course, if additional negative resonances are assumed the resonance parameters are more reasonable but then the whole analysis becomes disturbingly arbitrary.

In view of this it is not surprising that the interpretation of the cross section cannot avoid being indeterminate to some degree, and that the derived resonance parameters will depend on the initial assumptions. For example, Shore and Sailor[3] assumed that the lowest observable resonances at 0.28 and 1.14 eV interfere with each other but not with the negative level. Vogt[4] and Gordeev[5] also made this same assumption. On the other hand, Kirpichnikov *et al.*,[6] concluded that the cross section was better fitted if the 0.28- and 1.14-eV resonances were assumed to be in opposite spin states while the 1.14-eV resonance interfered with a level below the neutron binding energy. The latter assignment of opposite spins to the 0.28- and 1.14-eV resonances is supported by Dabbs, Walter, and Parker's[7] study of the directional distribution of fission fragments from aligned nuclei.

There are also other difficulties with the interpretation of the cross sections of the fissile nuclei. Using the formulas of multilevel theory, Lynn[8] has shown that when level widths are comparable to level spacings, the shape of the cross section becomes so distorted that a single peak can frequently be associated with several closely spaced levels. Lynn argues that this situation very probably occurs in all the common fissile nuclei. Evidence for this can be found in the discrepency between experimental and theoretical estimates of the effective number of fission channels. The collective-model description of the fission process, as developed by Bohr[9] and Wheeler,[10] invariably predicts a larger effective number of fission channels than is indicated by the experiments. For example, the predicted value for U^{235} is almost ten times the observed value. Lynn suggests that the discrepancy occurs because not all of the levels are correctly identified and accounted for in the analyses. Michaudon[11] has also reached this same conclusion from a statistical analysis of the observed resonance structure of the cross section.

While it is difficult to decide at this stage how serious an obstacle undetected resonances will ultimately be to an improved resonance analysis, nevertheless it is obvious that an experimental determination of the spin dependence of the cross section will eliminate at least one of the major sources of difficulty. With this idea in mind a program was initiated some years ago to study the interaction of polarized neutrons with a polarized U^{235} target. Not unexpectedly, this proved to be a formidable undertaking. Within the past two years, however, U^{235} targets have been developed which provide enough polarization to observe clearly the spin dependence of the cross section. Although some of the target parameters remain to be determined, the present measurements establish the spin dependence up to 2 eV sufficiently well to permit a substantially more reliable analysis of the cross section.

Aside from the resonance analysis, there is also interest in the more basic question of a possible correlation between the spins of the resonances and their fission widths. According to the collective model,[10] the $I-\frac{1}{2}=3^-$ levels of U^{236} should have an average fission width greater than the $I+\frac{1}{2}=4^-$ levels. Unfortunately, in the energy region below 2 eV there are not enough resonances to provide an adequate statistical sample, but when the measurements are extended to the region above 2 eV, it should be possible to test this prediction.

II. THEORY OF THE MEASUREMENT

A. Cross Section

Since the method used to determine the spin dependence of the cross section has been described in detail elsewhere,[12–14] we will only review it briefly here. For the interaction of polarized neutrons with polarized nuclei, the cross section takes the form

$$\sigma=\frac{I}{2I+1}(1-f_Nf_n)\sigma_- +\frac{I+1}{2I+1}\left(1+\frac{I}{I+1}f_Nf_n\right)\sigma_+ . \quad (1)$$

In this formula f_n and f_N are the neutron and nuclear polarizations while σ_- and σ_+ are the cross sections for

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[3] F. J. Shore and V. L. Sailor, Phys. Rev. **112**, 191 (1958).

[4] Erich Vogt, Phys. Rev. **112**, 203 (1958).

[5] I. V. Gordeev, At. Energ. USSR **12**, 408 (1962) [English transl.: Soviet J. At. Energy **12**, 433 (1962)].

[6] K. G. Ignat'ev, I. V. Kirpichnikov, and S. I. Sukhoruchkin, At. Energ. **16**, 110 (1964) [English transl.: Soviet J. At. Energy **16**, 121 (1964)]. I. V. Kirpichnikov, K. G. Ignat'ev, and S. I. Sukhoruchkin, At. Energ. **16**, 211 (1964). [English transl.: Soviet J. At. Energy **16**, 251 (1964)], Table I erroneously lists a bound level at −20 eV. The original paper lists the energy as −2.2 eV.

[7] J. W. T. Dabbs, F. J. Walter, G. W. Parker, Bull. Am. Phys. Soc. **8**, 69 (1963); and *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna 1965), Vol. 1, pp. 39–49.

[8] J. E. Lynn, Phys. Rev. Letters **13**, 412 (1964): *Nuclear Structure Study with Neutrons*, edited by M. Neve de Mevergnies, A. Assche, and J. Vervier (North-Holland Publishing Co., Amsterdam 1966), p. 125.

[9] A. Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva 1955* (United Nations, New York, 1956), Vol. 2, p. 151.

[10] J. A. Wheeler, *Fast Neutron Physics*, edited by J. B. Marion and J. L. Fowler, (Interscience Publishers, Inc., New York, 1963), Part II, p. 2051.

[11] A. Michaudon, doctoral thesis, University of Paris, 1964 (unpublished).

[12] H. Postma, H. Marshak, V. L. Sailor, J. F. Shore, and C. A. Reynolds, Phys. Rev. **126**, 979 (1962); H. Marshak, H. Postma, V. L. Sailor, F. J. Shore, and C. A. Reynolds, *ibid.* **128**, 1287 (1962).

[13] H. Postma, F. J. Shore, and C. A. Reynolds, Physica **30**, 713 (1964).

[14] F. J. Shore, C. A. Reynolds, V. L. Sailor, and G. Brunhart, Phys. Rev. **138**, B1361 (1965).

interaction in the states $J=I-\frac{1}{2}$ and $J=I+\frac{1}{2}$, respectively. It is advantageous to separate this expression into polarization-independent and polarization-dependent terms

$$\sigma=\sigma_0+f_N f_n \sigma_p\,, \tag{2}$$

where

$$\sigma_0=\frac{I+1}{2I+1}\sigma_+ +\frac{I}{2I+1}\sigma_- =\sigma_{I+1/2}+\sigma_{I-1/2} \tag{3}$$

and

$$\sigma_p=\frac{I}{2I+1}(\sigma_+-\sigma_-)=\frac{I}{I+1}\sigma_{I+1/2}-\sigma_{I-1/2}\,. \tag{4}$$

B. Transmission Effect

Expression (2) shows that the cross section will, in general, differ depending on the relative orientation of the neutron and nuclear polarization. Thus the transmission $\mathcal{T}_P$ through the target when the neutron beam is polarized parallel to the applied magnetic field H will be different from the transmission $\mathcal{T}_A$ when it is antiparallel to H. This is conveniently expressed by the *transmission effect*,

$$\mathcal{E}=(\mathcal{T}_P-\mathcal{T}_A)/(\mathcal{T}_P+\mathcal{T}_A)\,, \tag{5}$$

which is related to the polarization-dependent part of the cross section σ_P by the formula

$$\mathcal{E}\approx -\tfrac{1}{2}(1+\varphi)f_n \tanh[(\sigma_P/\sigma_0)f_N N\sigma_0 t]\,. \tag{6}$$

Since $(\sigma_p/\sigma_0)f_N N\sigma_0 t$ is usually $\ll 1$, it is a good approximation to write

$$\mathcal{E}\approx -\tfrac{1}{2}(1+\varphi)f_n(\sigma_p/\sigma_0)f_N N\sigma_0 t\,. \tag{7}$$

Of the terms not already defined, φ is the efficiency for reversing the beam polarization, N is the number of target nuclei per unit volume, and t is the target thickness. The quantity $\frac{1}{2}(1+\varphi)f_n$ can be determined independently, and $N\sigma_0 t$ is evaluated by measuring the neutron transmission through the unpolarized sample. Thus the experiment determines the product $(\sigma_p/\sigma_0)f_N$.

If the resonances are well enough separated so that there are energy regions where the cross section is essentially all in one or the other spin state, the quantity σ_p/σ_0 assumes either the value -1 or $I/(I+1)$, depending on whether the resonance has spin $I-\frac{1}{2}$ or $I+\frac{1}{2}$. Since in this case σ_p/σ_0 differs in magnitude as well as in sign, it is possible, when there are accessible resonances in both spin states, to make absolute spin assignments without knowing the sign of f_N.[13,14] This can be done by taking advantage of the fact that the differences in the magnitude of σ_p/σ_0 will be reflected in the relative magnitude of the transmission effect observed at individual resonances. Unfortunately, this procedure is impossible in U^{235} in the thermal region where the resonance structure is very poorly defined. It therefore becomes necessary to know the magnitude and sign of the nuclear polarization f_N to determine the magnitude and sign of σ_p/σ_0.

III. DESCRIPTION OF THE EXPERIMENT

A. Polarized Neutron Spectrometer and Associated Cryogenics

The apparatus used for the measurements consists of a polarized neutron crystal spectrometer with a target cryostat mounted on the spectrometer arm. The cryostat is designed to produce polarization by static methods and contains a two-stage adiabatic demagnetization refrigerator which will maintain targets in a magnetic field of up to 15 kOe at temperatures below 0.1°K for times in excess of 8h. Iron ammonium alum is used for both refrigerating salts. Superconducting Pb switches make thermal contact both between the upper salt and the 0.95°K liquid-helium bath, and between the two salts. The target is cooled by copper conductors connected to the lower paramagnetic salt. Additional detail concerning the apparatus can be found in Ref. 12.

Refrigerating salt temperatures are determined from ballistic-galvanometer measurements of the magnetic susceptibilities of the salts. These are converted from magnetic temperatures to absolute temperatures by using a scale calibrated by measurements of nuclear polarization in Re^{185}. These measurements have been previously described.[15]

B. Polarized U^{235} Targets

The production of significant nuclear polarization by means of static methods is usually dependent on finding a material with a large hyperfine interaction and with suitable magnetic characteristics. Although static methods have been successful on a variety of nuclei, U^{235} presents a number of problems which in combination make it a difficult material for a polarized target. For one thing, the nuclear moment of U^{235} is small and the spin is large; consequently a given field at the nucleus gives a comparatively small polarization. For another, specimens of U^{235} are hard to cool to low temperatures because of heating from α decay.[16] Finally, the magnetic properties of most uranium compounds have not yet been completely investigated, which makes the choice of a suitable chemical compound for the target very difficult. This last is a particularly serious handicap because both the direction and degree of nuclear polarization are intimately connected with the magnetic properties of the target system. To make absolute spin assignments for the U^{235} resonances, it is necessary to know the sign of the hyperfine interaction

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[15] L. Passell and R. I. Schermer, Phys. Rev. **150**, 146 (1966).

[16] The major part of the α activity normally associated with U^{235} specimens is actually due to U^{234} contamination. Target heating can be substantially reduced if U^{235} of high isotopic purity is available. In our most recent targets, which contain less than 0.1% U^{234}, the α heating due to U^{234} is only about twice that of U^{235}. Altogether, α heating amounts to 1-2 ergs/g sec in these targets. The enriched isotopes were obtained on loan from the U. S. Atomic Energy Agency, Division of Research, Stable Isotope Cross Section Research Pool.

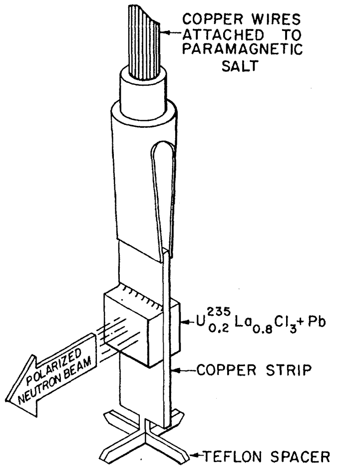

FIG. 1. Mounting arrangement for the $^{235}U_{0.2}La_{0.8}Cl_3$ target.

which defines the sign (direction) of the nuclear polarization f_N. In case the magnitude of σ_p/σ_0 is of interest it is necessary to determine the magnitude of f_N, which depends on the size of the hyperfine interaction, the degree of magnetic saturation of the target, and the temperature of the nuclear spin system. It is fortunate that the primary problem, the grouping of the resonances according to spin, does not require such detailed knowledge of the target properties and can be made simply on the basis of the sign of the observed transmission effects alone.

1. Magnetic Properties of the Target Materials

In the course of our efforts to develop polarized U^{235} targets, we have tried three different materials: pure uranium metal, the intermetallic compound UFe_2, and anhydrous UCl_3 dissolved in $LaCl_3$. There was evidence of some polarization in each of these targets, but only $U_xLa_{1-x}Cl_3$ produced sufficient polarization to allow measurements with good statistical accuracy.

The magnetic properties of these materials are discussed in detail in Appendix A. It will suffice to say here that UCl_3 orders antiferromagnetically at 20.3°K, UFe_2 ferromagnetically at 195°K, and that no sign of a localized magnetic moment has ever been observed in uranium metal.

2. Fabrication of the Targets

(*a*) *Uranium Metal.* The metallic uranium target, in the form of a slab, was sprayed with tin and soldered into a copper jacket so as to get good heat transfer at the interface. The copper jacket was soldered to a bundle of copper wires, the other ends of which were grown into the refrigerating salt. All soldered joints were located in regions of large magnetic fields to avoid superconducting interfaces. The target contained 10.9 g of metal and had dimensions $1.00\times0.75\times0.045$ in. The U^{234} content of this sample was only 0.1%, hence heating from α decay was small.

(*b*) *UFe_2 Intermetallic Compound.* The original UFe_2 target was a slab $1.01\times0.75\times0.13$ in. and weighed 20.7 g. It was mounted in the same manner as described above for the metal sample. Unfortunately, this specimen contained 1.1% U^{234}, which gave an estimated heating rate from α decay of 250 ergs/sec. Because of this high heat input and the assumed poor thermal conductivity of such an intermetallic compound, a second target was prepared from the same lot of UFe_2 in an effort to increase heat transfer. This target consisted of 20.3 g of finely powdered UFe_2 mixed with 20.4 g of Pb powder and pressed into a slab in a die press. The pressed slab was then coated with tin and mounted as above. Detailed procedures for preparing pressed powder targets have been described elsewhere.[17] The data showed no substantial difference between the two types of UFe_2 targets, i.e., no obvious improvement in heat removal from the pressed powder sample.

(*c*) *Uranium Trichloride.* Since $U_xLa_{1-x}Cl_3$ is a poor thermal conductor, it also was prepared as a pressed powder target, i.e., mixed with lead powder and pressed into slabs. The proportions were 22.6 g of $^{235}U_{0.2}La_{0.8}Cl_3$ to 22.4 g of lead. The mixed powders were formed into two slabs each of dimensions $1.01\times0.76\times0.32$ in., which were soldered to a copper strip as shown in Fig. 1.

The U^{3+} concentration in the trichloride targets was determined by chemical analysis. The material was also examined by x-ray analysis to make sure that it was free of inclusions of oxychloride or tetravalent uranium compounds.

The uranium concentration was selected by the criteria that $\mathcal{E}$ be maximized for a given thickness t. This occurs when the product $f_NN(U^{235})$ is maximized. Clearly the maximum $N(U^{235})$ which can be tolerated is less than the concentration at which antiferromagnetic spin cluster formation begins to cause a decrease in f_N. This maximum concentration was determined from magnetization measurements[18] made on samples of $^{238}U_xLa_{1-x}Cl_3$. These measurements indicated that at helium temperatures in moderate magnetic fields, approximate magnetic saturation occurs for $x\lesssim0.2$. It was on this basis that a 20% U^{235} concentration was chosen.[19] Possibly a somewhat higher concentration would have produced slightly larger transmission effects; however, it is very convenient in analyzing the data to have a substance whose magnetic properties do not change over the temperature range of the measurements as might occur if $x>0.2$.

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[17] R. I. Schermer and L. Passell, Rev. Sci. Instr. **36**, 709 (1965).

[18] These measurements were made by T. R. McGuire of the IBM Watson Research Center.

[19] The mixed salts, $^{235}U_{0.20}La_{0.80}Cl_3$ and $^{235}U_{0.02}La_{0.98}Cl_3$, were prepared by D. E. LaValle of the Oak Ridge National Laboratory.

Anticipating difficulties in interpreting the magnetic behavior of $^{235}U_{0.2}La_{0.8}Cl_3$, we also prepared two targets in which the U^{235} concentration was approximately ten times smaller, i.e., $^{235}U_{0.02}La_{0.98}Cl_3$. At such a low concentration the mixed salt should remain paramagnetic even at temperatures below 0.1°K. One of the targets consisted of $^{235}U_{0.0185}La_{0.982}Cl_3$ as a pressed powder-Pb mixture identical in form to the $^{235}U_{0.2}La_{0.8}Cl_3$ target. Since doubts as to the exact U^{235} concentration in this dilute target arose after the measurements were completed, a second dilute target was prepared in which the finely powdered salt was sealed into a brass capsule filled with He^4 gas under pressure. It was thought that better heat transfer could be achieved by this technique but the measurements showed no evidence that the second target reached a lower temperature. Although the results obtained with both targets were in excellent agreement we used only the data obtained with the second target in which the U^{235} concentration was more precisely determined.

Finally, we prepared a target of $U_{0.2}La_{0.8}Cl_3$ which was essentially identical to the $^{235}U_{0.2}La_{0.8}Cl_3$ target except that uranium of normal isotopic composition, i.e., 0.7% U^{235}, was used. The purpose of this target was to provide a direct measurement of the contributions of La and Cl to the observed transmission effects.

IV. MEASUREMENTS AND DATA REDUCTION

The quantity of interest is the transmission effect due to the U^{235} component of the target. While this is obtained directly from the data on U metal, there are complications for both UFe_2 and $U_xLa_{1-x}Cl_3$. First, since UFe_2 and $U_xLa_{1-x}Cl$ are magnetic, there is an "electronic" effect[20] which arises from the interference between nuclear coherent scattering and scattering from the magnetic electrons. This effect is temperature-independent and thus may be distinguished from the temperature-dependent nuclear effect. Further, in the $U_xLa_{1-x}Cl_3$ targets, there is also a "background" transmission effect from the La and Cl nuclei which become partially polarized along with the U^{235}. A separate measurement must be made to correct for these contributions. Finally, to intercompare data taken with different targets there are corrections for differences in the target thickness Nt and target temperature. Taken altogether, these corrections become quite complicated, especially for the trichloride targets, and cannot be made in a completely rigorous way.

A. Measurements

Table I summarizes the measurements made with each target. The neutron energy and the range of the refrigerating salt temperature are given for each entry.

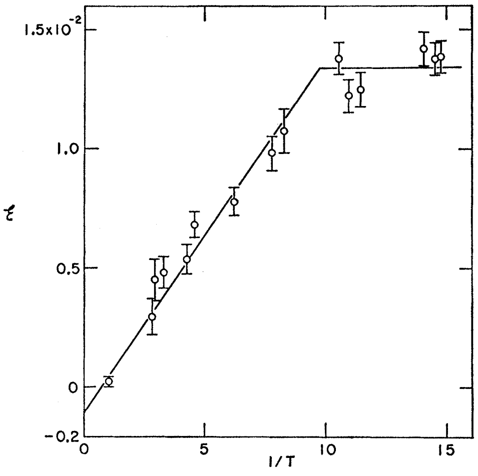

FIG. 2. The temperature dependence of the transmission effect $\mathcal{E}$ in $^{235}U_{0.2}La_{0.8}Cl_3$ as observed at 0.075 eV. The break in the curve indicates lack of thermal contact at the lowest temperatures.

B. Data Reduction

1. *Separation of Nuclear and Electronic Effects*

After adiabatic demagnetization, the refrigerating salt temperature rises slowly, and $\mathcal{E}$, which is the sum of the temperature-independent electronic effect and the temperature-dependent nuclear effect, will slowly change as the target temperature increases. To separate the contributions of the two terms the data were fitted to an expression of the form

$$\begin{aligned}\mathcal{E}(\text{observed}) &= \mathcal{E}(\text{electronic}) + \mathcal{E}(\text{nuclear})\\ &= \alpha + \beta/T\,, \end{aligned} \tag{8}$$

where α and β are constants. In uranium metal, which is nonmagnetic, it was anticipated that $\alpha \equiv 0$, and this is supported by the data. In magnetic materials $\alpha \neq 0$ at low neutron energies, but it is expected that α will decrease rapidly with energy. This behavior was observed

TABLE I. Range of the measurements.

Target	Neutron energy (eV)	Temp. range
^{235}U metal	0.075	$T = 1°K$ and $T \lesssim 0.18°K$
$^{235}UFe_2$	0.078	$T < 1°K$
	0.115	$T < 1°K$
	0.275	$T < 1°K$
$^{235}U_{0.2}La_{0.8}Cl_3$	0.075	$0.070 \lesssim T \lesssim 1°K$
	0.115	$T = 1°K$ and $T \lesssim 0.15°K$
	0.275	$T = 1°K$ and $T \lesssim 0.15°K$
	0.364	$T \lesssim 0.15°K$
	0.584	$T \lesssim 0.15°K$
	0.835	$T \lesssim 0.15°K$
	1.14	$T \lesssim 0.15°K$
	2.04	$T \lesssim 0.15°K$
$U_{0.2}La_{0.8}Cl_3$ (normal isotopic abundance)	0.075	$T = 1°K$ and $T \lesssim 0.1°K$
	0.275	$T = 1°K$ and $T \lesssim 0.1°K$
	1.14	$T = 1°K$ and $T \lesssim 0.1°K$
$^{235}U_{0.02}La_{0.98}Cl_3$	0.075	$T = 1°K$ and $T \lesssim 0.2°K$

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[20] R. I. Schermer, Phys. Rev. **130**, 1907 (1963), Eq. (17).

TABLE II. The transmission effect (in percent) as observed in various targets. Listed at each energy are the neutron polarization and the filter(s) used to remove second-order neutrons from the beam. The values for $^{235}U^{3+}$ are derived by subtracting from the observed values of $\mathcal{E}$ the electronic effect and the contributions of the La and Cl.

Target	T degrees Kelvin	$N(U)t$ 10^{-3} atoms/bn	0.075 eV Pu^{239} filter $f_n=0.84$	0.115 eV Eu filter $f_n=0.85$	0.275 eV Pu^{240} filter $f_n=0.85$	0.364 eV In filter $f_n=0.86$	0.584 eV Te filter $f_n=0.87$	0.835 eV Tb,Cd filters $f_n=0.88$	1.14 eV Ta,Cd filters $f_n=0.90$	2.04 eV Sm,Cd filters $f_n=0.90$
^{235}U metal	0.10	5.77	0.046±0.012							
^{235}U Fe$_2$	?	6.88	−0.333±0.090[a]	−0.198±0.069	0.089±0.040					
$^{235}U_{0.2}La_{0.8}Cl_3$	0.10	2.23	1.344±0.026	1.027±0.032	−0.416±0.079	−0.112±0.051	0.131±0.058	0.173±0.063	0.676±0.065	−0.087±0.043
$U_{0.2}La_{0.8}Cl_3$[b]	0.10	2.20	−0.110±0.028	−0.030±0.016	−0.023±0.019	−0.021±0.021	−0.018±0.025	−0.017±0.028	−0.016±0.029	−0.014±0.033
$^{235}U^{3+}_{0.2}$	0.10	2.23	1.454±0.038	1.057±0.036	−0.393±0.081	−0.091±0.055	0.149±0.063	0.190±0.069	0.692±0.071	−0.073±0.054
$^{235}U_{0.02}La_{0.98}Cl_3$	0.10	0.255	0.073±0.012							
$^{235}U^{3+}_{0.02}$	0.10	0.255	0.107±0.022$<\mathcal{E}<$ 0.115±0.025							

[a] This measurement was made at 0.078 eV.

[b] Measurements were made at 0.075, 0.275, and 1.14 eV. Values at other energies are interpolated.

in $U_{0.2}La_{0.8}Cl_3$, where measurements were made over an extended range of energies. At 0.275 eV, α was too small to be measured and therefore it was assumed that $\alpha=0$ at this and all higher energies for all three trichloride targets.

2. *Reduction to a Standard Temperature*

It is essential that the target be in good thermal contact with the refrigerating salt so that the temperature of the salt, which is the measured quantity, is also the temperature of the target. This is believed to be the case in the dilute and normal uranium trichloride targets, which had low-α heating and were otherwise very similar to targets we have used in other experiments.[17] In uranium metal there was more α heating but presumably the heat ransfer was also better; hence the assumption of thermal equilibrium is probably not unreasonable. The UFe_2 target, on the other hand, was almost certainly much warmer than the refrigerating salt because of severe α heating.

In the $^{235}U_{0.2}La_{0.8}Cl_3$ target it was possible to study in detail the variation of $\mathcal{E}$ with temperature, since at 0.075 eV, $\mathcal{E}$ could be determined very accurately. The results of these measurements are shown in Fig. 2. The electronic effect shows up as the negative intercept at $1/T=0$; the sharp break at 0.1°K indicates that because of α heating the target did not cool below this temperature although the refrigerating salt got much colder. Therefore, in analyzing all data for $T<0.1$°K we assumed the $^{235}U_{0.2}La_{0.8}Cl_3$ target was at a constant temperature and simply averaged the data as indicated by the horizontal straight line through the low-temperature points. For $T>0.12$°K we assumed the target and refrigerating salt to be in thermal equilibrium and applied Eq. (8). The intersection of the two fitted lines in Fig. 2 at 0.102°K was chosen as the standard temperature to which all values of $\mathcal{E}$ were subsequently normalized.

It should be noted that in comparing data taken with the same target at different energies, uncertainties in the target temperature will not significantly influence the results. But to minimize the possibility of error, all data were nevertheless taken over the same temperature range at each energy.

However, in intercomparing data taken with different targets, or when comparing observations with theory, the determination of the target temperature remains of great importance. It is certainly a major source of uncertainty in attempts to estimate the nuclear polarization f_N.

3. *Variation in Effective Target Thickness*

To intercompare results obtained with different trichloride targets it is also necessary to account for the variation of the product Nt from target to target. Since $\mathcal{E}$ is proportional to Nt the correction is quite straightforward. As a matter of convenience, all the trichloride measurements were normalized to the same number of Cl atoms/cm^2.

4. Background Corrections for the Trichloride Targets

There remains the question of determining the contributions of La and Cl to the observed transmission effects. All such corrections were based on measurements made with a $U_{0.2}La_{0.8}Cl_3$ target containing uranium of normal isotopic composition. The details of these corrections are given in Appendix B.

C. Results and Comments

The results of the measurements normalized to a standard temperature and adjusted for target thickness variations are given in Table II. Listed are the measured values of $\mathcal{E}$ for each target and also the values obtained for $^{235}U^{3+}{}_{0.2}$ and $^{235}U^{3+}{}_{0.02}$ alone. The latter were derived by subtracting from the observed $\mathcal{E}$'s the electronic, La, and Cl contributions.

It is evident from inspection of the table that $\mathcal{E}$ in UFe_2 is proportional, but of the opposite sign, to $\mathcal{E}$ in either $U_{0.2}La_{0.8}Cl_3$ or uranium metal. This indicates that there is a negative hyperfine field in UFe_2. Unfortunately the temperature of this target could not be determined with any accuracy hence it was not possible to obtain a reliable estimate of the magnitude of the field.

A second point of interest is that within the accuracy of the data, $\mathcal{E}$ in the $^{235}U_{0.02}La_{0.98}Cl_3$ target is $\frac{1}{10}$ of $\mathcal{E}$ in the $^{235}U_{0.2}La_{0.8}Cl_3$ target. Thus there appears to be no indication of magnetic coupling between uranium ions in this concentration range.

D. Calculation of $(\sigma_p/\sigma_0)f_N$ from $\mathcal{E}$

If the cross section does not vary rapidly with energy, the product $(\sigma_p/\sigma_0)f_N$ can be determined by substituting the observed values of $\mathcal{E}$, $f_n(1+\varphi)/2$ and $N\sigma_0 t$ directly into Eq. (7). In the neighborhood of a resonance, however, it is necessary to take into account the effects of spectrometer resolution, which become quite pronounced. In the interest of brevity we will not describe the method used to make the required resolution corrections since it is discussed in detail in Refs. 13 and 14. The values of $(\sigma_p/\sigma_0)f_N$ derived from the data obtained with the $^{235}U_{0.2}La_{0.8}Cl_3$ target have been listed in Table III. They have also been plotted in Fig. 3.

Resolution corrections were necessary at both 0.275 and 1.14 eV. These corrections depend to some extent on how the cross section is analyzed into its component parts, hence $(\sigma_p/\sigma_0)f_N$ at the resonance energies could be systematically in error by perhaps as much as 10%. At 2.04 eV possible systematic errors in the resolution correction, when combined with the large uncertainty in the observed value of $\mathcal{E}$ itself, make the value of $(\sigma_p/\sigma_0)f_N$ of very doubtful reliability. Therefore only the sign, but not the magnitude, is included in the tabulation.

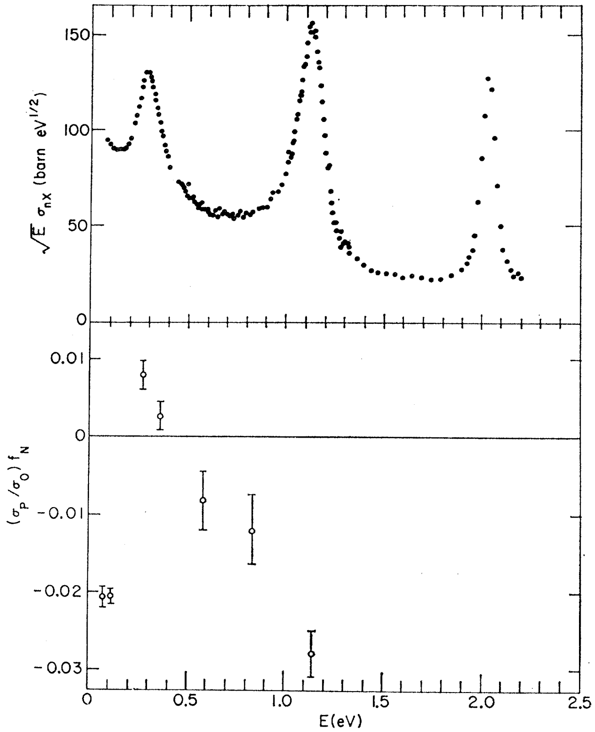

FIG. 3. The measured values of $(\sigma_p/\sigma_0)f_N$. For convenience of comparison with the cross section, the data of Shore and Sailor (Ref. 3) is plotted as the product $(\sqrt{E})\sigma_{\text{total}}$ in the upper part of the figure on the same energy scale. The value obtained at 2.04 eV is not plotted because it is of questionable reliability; however, the data indicate that the resonance is probably in the same spin state as the 0.275-eV resonance.

V. DISCUSSION

There are certain conclusions concerning the spin dependence of the cross section which are obvious from inspection of either Fig. 3 or Table III.

(i) The 0.275- and 1.14-eV resonances are in opposite spin states.

(ii) The thermal cross section is primarily in the same spin state as the 1.14-eV resonance.

TABLE III. Values of $(\sigma_p/\sigma_0)f_N$ derived from the data obtained with $^{235}U_{0.2}La_{0.8}Cl_3$.

Neutron energy (eV)	$(\sigma_p/\sigma_0)f_N$
0.075	$-2.07\pm0.14\times10^{-2}$
0.115	-2.06 ± 0.10
0.275	$+0.80\pm0.18$
0.364	$+0.26\pm0.18$
0.584	-0.82 ± 0.38
0.835	-1.20 ± 0.48
1.14	-2.78 ± 0.30
2.04	+

(iii) The 2.08-eV resonance is probably in the same spin state as the 0.275-eV resonance. This conclusion is not, however, as well established statistically as the others.

If we assume, following the discussion in Appendix A, that U^{235} has a negative nuclear magnetic moment and therefore f_N is negative, then we can further state that

(i) $J=I+\frac{1}{2}=4$ for the 1.14-eV resonance and for at least the predominant part of the thermal cross section.

(ii) $J=I-\frac{1}{2}=3$ for the 0.275-eV resonance and probably for the 2.08-eV resonance as well.

So far as we can determine, the only other experiment bearing on this problem is Dabbs, Walter, and Parker's study[7] of the directional distribution of fission fragments from aligned nuclei. Their measurements indicated that the 0.275- and 1.14-eV resonances are in opposite spin states, in agreement with our own conclusions.

We are not aware of any analysis of the U^{235} cross section which is completely consistent with all of the observations listed above. The closest is probably that of Kirpichnikov *et al.*,[6] who assigned opposite spins to the 0.275- and 1.14-eV resonances. But they also postulated that there is only one negative resonance and that this resonance is in the same spin state as the 1.14-eV resonance and interferes with it. They considered the 0.275- and 2.08-eV resonances to be isolated, noninterfering resonances. If these assumptions are correct, then the product $(\sigma_p/\sigma_0)f_N$ should have the same value at all energies below 1.14 eV except in the near neighborhood of 0.275 eV, which is the only region where this resonance contributes significantly to the cross section. Inspection of Fig. 3 shows that this is not what is observed. The value of $(\sigma_0/\sigma_p)f_N$ at 0.075 and 0.115 eV is about twice that at 0.584 and 0.835 eV.

Of course it is implicitly assumed in this argument that the peaks in the cross section are associated with single levels of U^{235}. As long as this assumption is retained, it is not possible to account for the observed variation of σ_p/σ_0 with energy unless it is also assumed that negative resonances in both spin states contribute to the thermal cross section. This is an attractive assumption because it also offers a more reasonable explanation for the large thermal cross section.

Additional evidence that the thermal cross section is a mixture of spin states can be provided by calculating f_N and thus arriving at an estimate of σ_p/σ_0. Although there are questions concerning the La and Cl contributions, the temperature, and the magnetic properties of the targets (see Appendices A and B), we have reason to believe that the calculated values of f_N are correct at least to their order of magnitude. In support of this we note that at 0.075 eV, where measurements were made with U^{235} metal and both dilute and concentrated trichloride targets, the derived values of σ_p/σ_0 are in rough agreement:

$$\begin{aligned}
&{}^{235}U^{3+}{}_{0.2} && \sigma_p/\sigma_0=0.187\pm0.005\\
&{}^{235}U^{3+}{}_{0.02} && \sigma_p/\sigma_0=0.13\pm0.03\\
&{}^{235}U\text{ metal} && \sigma_p/\sigma_0=0.32\pm0.08\,.
\end{aligned}$$

Taking the above values of σ_p/σ_0 and using Eqs. (3) and (4) we find

$$\begin{aligned}
&{}^{235}U^{3+}{}_{0.2} && \sigma_{I+1/2}/\sigma_0=0.67\,,\\
&{}^{235}U^{3+}{}_{0.02} && \sigma_{I+1/2}/\sigma_0=0.64\,,\\
&{}^{235}U\text{ metal} && \sigma_{I+1/2}/\sigma_0=0.74\,,
\end{aligned}$$

i.e., about two thirds of the cross section at 0.075 eV is in the $I+\frac{1}{2}$ state, which is consistent with our earlier conclusion.

According to the collective model of the fission process,[10] the $I-\frac{1}{2}=3^-$ levels should have on the average a greater fission width than the $I+\frac{1}{2}=4^-$ levels. Unfortunately, only three resonances were examined in this experiment and this obviously is not an adequate statistical sample. But even if more resonances were studied it is doubtful that the resonance parameters are sufficiently well determined at the moment to provide a reliable test. Clearly, all of the existing multilevel analyses are based at least in part on incorrect assumptions. An analysis which is consistent with the presently observed spin dependence of the cross section might very well yield considerably different values for the resonance parameters. Possibly it might also throw new light on the question of "missed" resonances as raised by Lynn and Michaudon. In the absence of such an analysis, however, there is one experimental observation which may have some relevance; namely, that the values of σ_p/σ_0 obtained by estimating the nuclear polarization indicate that nowhere, even at the peak of the 1.14-eV resonance, is the cross section entirely in one spin state.

Finally, with regard to hyperfine interactions in the materials studied, our measurements indicate a negative hyperfine field at the uranium nucleus in UFe_2. There was no evidence of a hyperfine interaction in U^{235} metal.

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ation of the cryostat was rendered by C. J. Crookes. Their help is gratefully acknowledged.

APPENDIX A. MAGNETIC PROPERTIES OF THE TARGET MATERIALS

1. *Uranium Metal.* Uranium metal shows no indication of either localized magnetic moments or long-range magnetic order down to 2°K.[21] A large specific heat has been found in U^{235} in the temperature range 0.3–0.7°K,[22] but since there is no evidence of any atomic magnetic moment this is probably due to an electric quadrupole interaction rather than a magnetic hyperfine interaction. The very small transmission effect observed in our measurements confirms this.

2. *UFe_2.* There are definite indications of ferromagnetic ordering in UFe_2 at temperatures below 195°K.[23] However, the most recent magnetic measurements[24] indicate that the moment is localized on the Fe sites and that there is little or no moment associated with the U atoms. Nothing is known as to what, if any, magnetic hyperfine field exists at the U nucleus. The transmission effect in UFe_2, while small, was found to be significantly larger and of opposite sign to that observed in uranium metal. It seems, therefore, that there is a hyperfine interaction in this material but we were not able to make any quantitative estimate of its magnitude.

3. *UCl_3.* A considerable amount of information is available on both the hyperfine interaction in anhydrous UCl_3 and the magnetic properties of this material. Susceptibility measurements[25] made with dilute solutions of UCl_3 in $LaCl_3$ are consistent with the expected ${}^4I_{9/2}$ ground-state characteristic of three unpaired $5f$ electrons. There are also measurements[26] of paramagnetic resonance absorption in $LaCl_3$ crystals containing 2 mole % UCl_3 which support this conclusion.

The susceptibility measurements also showed that concentrated UCl_3 orders antiferromagnetically at 20.3°K. Thus, unless there is a very large field available to break down the antiferromagnetic ordering, UCl_3 can only be used for a polarized target in magnetically dilute form, i.e., mixed with a diamagnetic isomorphous compound such as $LaCl_3$. Depending upon the dilution, some fraction of the U^{3+} ions behave as essentially free paramagnetic ions; the remainder form magnetic clusters of various types. Unfortunately, the magnetic behavior of systems of such complexity cannot be properly interpreted without a detailed understanding of the couplings between the various pairs of ions. While electron spin resonance measurements[27] have been made on Nd^{3+} (which has the same electronic structure as U^{3+}) in the form of solutions of $NdCl_3$ in $LaCl_3$, nothing comparable has as yet been done with U^{3+}.

Hyperfine structure has been observed in the paramagnetic resonance absorption spectrum of 2 mole % UCl_3 in $LaCl_3$.[26] The absolute magnitudes of the hyperfine interaction constants were determined from the data, but the experiment did not give directly the sign of the hyperfine interaction. Because of these measurements it is possible to estimate the magnitude of the nuclear polarization in UCl_3 quite accurately. According to Ref. 26

$$g_{\parallel}=4.153\,,\quad g_{\perp}=1.520\,,$$

and

$$A=0.0176\ \text{cm}^{-1},\quad B=0.00568\ \text{cm}^{-1}.$$

The formula for calculating the average nuclear polarization in a powdered sample (i.e., the angle averaged projection of f_N on the external field direction) has been given elsewhere[28];

$$f_N=\frac{I+1}{6KT}[\mathrm{A}g_{\parallel}R_2+\mathrm{B}g_{\perp}(R_0-R_2)]\,, \tag{A1}$$

where

$$R_n=\int_0^{\pi/2}\left(\frac{1}{g}\right)\tanh\left(\frac{g\beta H}{2KT}\right)\cos^n\theta\,\sin\theta d\theta$$

and

$$g^2=g_{\parallel}^2\cos^2\theta+g_{\perp}^2\sin^2\theta\,.$$

Using the above constants

$$f_N=0.0113/T\,. \tag{A2}$$

Determining the sign of f_N is less straightforward. First, to find the sign of the hyperfine field for $U^{3+}(5f^3)$ in the anhydrous trichloride, there is the theory of Elliott and Stevens,[29] which was developed for Nd^{3+} in the ethyl sulfate. In both ions the ground state is ${}^4I_{9/2}$ and the point symmetry is C_{3h}.[26] The ground state is split into five Kramers doublets, the lowest of which is given by

$$\mathrm{cox}\theta\,|J_z=\pm\tfrac{7}{2}>+\sin\theta\,|J_z=\mp\tfrac{5}{2}>\,,$$

with $\theta\approx 17°$. Admixture of other J values is small as shown by the fact that the ratio $\mathrm{A}g_{\perp}/\mathrm{B}g_{\parallel}=1.13$ is close to unity, as is also the case with Nd^{3+}. In this circumstance, it follows from formulas (4.7)–(4.10) of Ref. 29 that the signs of ($\mathrm{A}g_{\parallel}$) and ($\mathrm{B}g_{\perp}$) in Eq. (A1), and thus the sign of f_N, are given solely by the sign of the nuclear gyromagnetic ratio g_N.

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In the above argument core polarization has been neglected. Note, however, that in the first half of the $4f$ shell, in which S is antiparallel to J, one expects H_C, the effective field due to core polarization, to be positive.[30] There are no direct calculations for $5f$ ions, but in passing from $3d$ to $4d$ ions H_C is found to increase approximately threefold with no change in sign.[30] It seems likely that the same thing will happen when going from $4f$ to $5f$.

Even if H_C were negative, it is doubtful that it could be large enough to override the positive effective field H_E due to the $5f$ shell alone. For Nd^{3+}, $H_C=-90(g-1)\times J=+300$ kOe.[30] By extrapolation it might be expected that for U^{3+}, $H_C\approx+1000$ kOe. From the theory of Elliott and Stevens for U^{3+} with $\langle J_z\rangle=J=\frac{9}{2}$, $H_E\approx+5300$ kOe, i.e., $H_E\gg H_C$. Thus it is reasonable to assume that the sign of the nuclear polarization is determined solely by the sign of g_N.

Unfortunately there have been no direct measurements of the sign of g_N, although the collective model calculations of Mottelson and Nilsson[31] predict that g_N is negative. Van der Sluis and McNally[32] have made optical hyperfine measurements on U^{235} and have stated that g_N is negative; however, the basis of this assignment is not readily apparent since the electronic structure of the level producing the hyperfine splitting is not known. In fact Blaise *et al.*[33] did not reach any conclusion as to the sign of g_N from an identical measurement. On the basis of the Mottelson-Nilsson theory we shall assume that $g_N<0$ and hence $f_N<0$. Since the absolute spin assignments for the resonances are based on this choice of sign, it is clear that a direct experimental determination of the sign of g_N would be very valuable.

APPENDIX B. "BACKGROUND" CORRECTIONS FOR LA AND CL IN THE TRICHLORIDE TARGETS

Assuming that the observed transmission effects in the three trichloride targets have been adjusted to the same temperature and target thickness then $\mathcal{E}$ can be expressed in the following form:

$$\mathcal{E}(20\%)=\mathcal{E}({}^{235}\mathrm{U}_{0.2})+\mathcal{E}(\mathrm{La}_{0.8})+\mathcal{E}(\mathrm{Cl}_3)+\mathcal{E}(\mathrm{Electronic})\,, \quad \text{(B1a)}$$

$$\mathcal{E}(2\%)=\mathcal{E}({}^{235}\mathrm{U}_{0.02})+\mathcal{E}(\mathrm{La}_{0.98})+\mathcal{E}(\mathrm{Cl}_3)+0.1\mathcal{E}(\mathrm{Electronic})\,, \quad \text{(B1b)}$$

and

$$\mathcal{E}(\mathrm{norm})=\mathcal{E}(\mathrm{La}_{0.8})+\mathcal{E}(\mathrm{Cl}_3)+\mathcal{E}(\mathrm{Electronic})\,. \quad \text{(B1c)}$$

The evaluation of the electronic term has been discussed in Sec. IV B1. What remains is to estimate the contributions of the La and Cl.

Formula (B1c) gives the proper background correction for (B1a), except that $\mathcal{E}$(norm) was not measured at as many energies as $\mathcal{E}(20\%)$. The electronic contribution was therefore subtracted from (B1c) and the energy dependence of the remainder fitted to an expression of the form $\gamma+\delta E^{-1/2}$, where γ represents the effect of scattering and δ of absorption.[34] The resulting interpolated values of $\mathcal{E}$(norm) were then subtracted from $\mathcal{E}(20\%)$, to yield the transmission effects due to ${}^{235}\mathrm{U}^{3+}{}_{0.2}$ alone, given in Table II.

It is clear that (B1c) is *not* the correct background for (B1b), even after subtracting off the electronic component. Taking the value listed in Table II at 0.075 eV and 0.10°K and subtracting off the electronic contribution we have

$$\mathcal{E}(\mathrm{La}_{0.8})+\mathcal{E}(\mathrm{Cl}_3)=-0.031\pm0.016\times10^{-2}\,. \quad \text{(B2)}$$

Assuming brute force polarization of La and Cl this may be expressed as a linear relation between (σ_p/σ_0) for La and (σ_p/σ_0) for Cl. By further assuming that all of the observed $\mathcal{E}$ is due to either the La alone or to the Cl alone we can arrive at upper and lower limits on the background correction for the dilute target:

$$-0.042\pm0.022\times10^{-2}\leq\mathcal{E}(\mathrm{La}_{0.98})+\mathcal{E}(\mathrm{Cl}_3)\leq-0.034\pm0.018\times10^{-2}\,. \quad \text{(B3)}$$

Finally, inserting (B3) into (B1b) we obtain upper and lower bounds to the transmission effect in the 2% sample due to the ${}^{235}\mathrm{U}^{3+}{}_{0.02}$ acting alone, as given in Table II.

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[34] We are assuming that there are no nearby resonances in either La or Cl. Actually, a very weak resonance has been observed in La at 0.75 eV [J. A. Harvey, R. C. Block, and G. G. Slaughter, Bull. Am. Phys. Soc. **4**, 385 (1959)] but its contribution to the total cross section is extremely small.