Neutron Resonance Spectroscopy. XIII. Na to 320 keV*

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Results are presented of measurements of the total cross section vs energy which was measured to 320 keV for pure metallic sodium. R-matrix fits were made to the seven prominent observed resonances. The best-fit parameters for (E_0, Γ, J, l) for each resonance are: (2.850, 0.400, 1, 0); (53.15, 1.08, 2, 1); (199.0, 4.5, 1, 0); (212.5, 15, 0, 1); (238, 3.9, 2, 1); (242, 3.5, 0, 1); (297, 1.8, 2, 0), where E_0 and Γ are in keV. Three levels are assigned to be s levels, and four to be p levels. This gives $10^4 S_0 = 0.24\ (1^{+1.91}_{-0.58})$ and $10^4 S_1 = 2.06(1^{+0.77}_{-0.39})$. The S_1 evaluation includes the presence of weak p levels seen in capture measurements by others.

I. INTRODUCTION

This paper reports results of high-resolution neutron-spectroscopy measurements of the total cross section of Na to 320 keV. It is one of a series[1-12] reporting results of time-of-flight (t.o.f.) resonance neutron spectroscopy using the Columbia University Nevis synchrocyclotron. In addition to the σ_t vs E cross-section behavior, we give the results of R-matrix fits to seven Na resonances which we observe in this energy region. From the viewpoint of nuclear physics, sodium, $A = 23$, is in a mass region where the s-wave strength function, S_0, is relatively low, but the p-strength function, S_1, is important. Three of the resonances are best fitted as s levels and four as p levels.

The total neutron cross section of Na is of great importance in fast-breeder-reactor applications, where liquid Na is used as a coolant. Of special interest is the peak cross section and spin and parity of the level at 2.85 keV, for which conflicting assignments exist. A recent evaluation of the ENDF/B sodium data set by Paik and Pitterle[13] at Westinghouse shows that the uncertainties in the sodium cross sections are important in the calculation[13] of the effective multiplication factor k_{eff} and the Doppler void coefficients for fast reactors. Older cross-section measurements by Whalen and Smith[14] at Argonne National Laboratory (ANL) span the energy region 100 to 300 keV, however no resonance parameters were given. Recently, several new measurements have been made, most notably the total cross-section results of Nebe and Kirouac at Karlsruhe[15] ranging from ~300 keV to several MeV and the capture results of Hockenbury *et al.*[16] at Rennsalaer Polytechnical Institute (RPI) below a few hundred keV. Our results extend from 10 eV up to overlap the low-energy end of the Karlsruhe data for the s level at 297 keV. They complement the capture results obtained at RPI.

Data for the results given here were obtained during two major runs using the synchrocyclotron. The results are all from transmission measurements using our 202.05-m flight path. Details of the over-all system and analysis methods below ~20-keV energy have been given in Ref. 8 for the earlier run, in Ref. 10 for the later run, and in earlier papers referred to in these references. During the earlier run, we had transmission measurements using metallic Na samples having $1/n$ values of 4.8, 30, 139, and 550 b/atom. Preliminary results have been reported below ~100 keV for those measurements.[17,18] The proper treatment of the energy and sample-dependent background (B.G.) and of the B. G. for open beam to obtain the B.G.-subtracted open rate above 20 keV is much more difficult than below 20 keV. Such corrections are essential to obtain correct sample transmission (T) and total cross section (σ) values.

During the later run, we used high-purity metallic Na samples in transmission having $1/n$ values of 5.25 and 24 b/atom. Of particular interest was the inclusion of "standard filter" measurements to help us evaluate the necessary background subtractions, etc., needed to obtain reliable (T, σ) above ~20 keV. This method, described in more detail below, gave a set of reliable σ values for Na at many energies over the energy region, at energies between those of the Na resonances. This information was used to reprocess the earlier data for Na which provides almost all of the σ vs E results given in this paper. The only exception is that the later data for the $1/n = 24$ b/atom Na sample was used in the region of the 53-keV resonance.

II. EXPERIMENTAL DETAILS AND ANALYSIS PROCEDURES

The metallic Na samples were sealed in polyethylene bags having ~0.1-mm wall thickness.

Corrections were made for the known thicknesses of polyethylene in the beam during the transmission measurements. Inspection of the data indicates that the resolution was about 0.3 nsec/m over most of the energy region. The levels below 180 keV were much wider than the resolution width or the Doppler width. The resolution limited the peak measured σ to values below the true peaks for three levels above 100 keV. Examples of the standard filter measurements are shown in Figs. 1 and 2, and the results are given in Table I. Three different standard filters were used in the beam and measurements made with and without the thick Na sample for each standard filter. The filters were composed of 7.6-cm-thick Fe, of 1.27-cm-thick Co, and 1.27-cm-thick Cu (metal filters). The transmission spectra through the Co and the Cu filters show pronounced transmission dips at their strong resonances, plus their energy-dependent backgrounds. When the $1/n = 5.25$ b/atom Na sample was also in the beam, the magnitude of these dips was reduced by the energy-dependent transmission of the Na sample. For the Co and Cu dips not near the Na levels, this permitted evaluation of (T, σ) values for the $1/n = 5.25$ b/atom Na sample at these Co and Cu resonance energies. When the thick Fe filter was present, the structure in the spectrum was mainly due to the transmission peaks on the low side of the strong ^{56}Fe s levels where there is a near cancellation of the Fe scattering cross section.[19]

In Fig. 1, the transmission, in arbitrary units, is shown for the Fe filter alone (below), and the corresponding result with the Na also present is shown displaced upward an arbitrary amount.

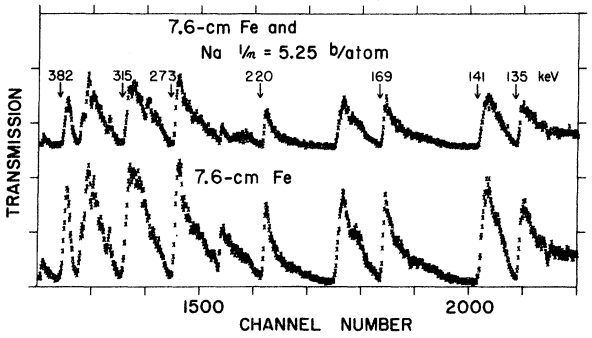

FIG. 1. A portion of the relative transmission vs energy is presented using 7.6-cm iron as a standard filter in the neutron beam. Background subtractions have not been made. The lower curve is for the Fe filter alone, while the (arbitrarily displaced) upper is for the case where our $1/n = 5.25$ b/atom Na sample is also present. The Na sample transmission is the factor by which the structure amplitude, Fe alone, is reduced when Na is also present, if there are no Na levels in the region.

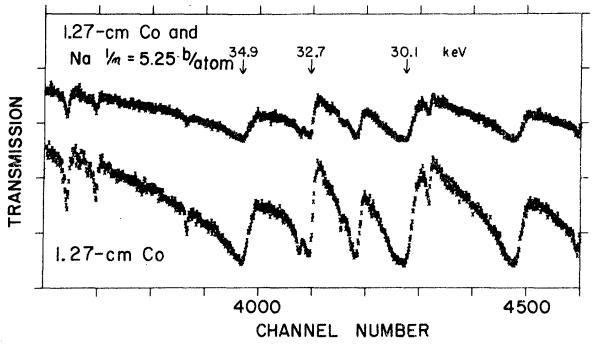

FIG. 2. Similar results to those of Fig. 1, but using a 1.27-cm-thick cobalt standard filter sample, with and without the thick Na sample.

Figure 2 illustrates a portion of the spectra using the Co filter, with and without the Na sample. Some of the resulting sodium total cross-section values and evaluation energies are given in Table I.

The statistical uncertainty of these values is indicated by their random fluctuations from one another in regions not near those of the Na resonances.

The measured σ vs E values for Na over the energy region to 320 keV are mainly from the thick Na sample data. This sample has good bottoming transmission dips ($T = 0$) at the 2.85- and 53-keV resonances where the background subtraction for

TABLE I. Na cross sections resulting from the standard filter measurements.

E_0 (keV)	σ_t (b)	E_0 (keV)	σ_t (b)
Using the iron filter			
382	3.18	169	2.88
315	3.08	141	3.43
273	2.52	135	3.31
220	3.33	83	3.50
182	3.28	74	3.83
Using the cobalt filter			
92	3.41	41	4.02
89	3.63	34	4.29
79	3.77	29.6	3.94
77	3.51	29	3.84
72	3.80	22	4.01
59	4.64		
57	4.98		
46	4.21		
Using the copper filter			
42.0	4.16	10.7	5.29
33.4	3.92	9.8	5.01
29.5	4.05	9.2	5.63
22.8	4.29	7.9	6.27
21.0	4.05	4.8	11.2

this sample is thus established. A background for that sample over the entire energy region is then established, essentially as described in Ref. 7. Since the B. G.-subtracted count vs E for the thick sample is known, and the sample T is known at the energies listed in Table I, the implied B. G.-subtracted count at these energies is also known for the thinner Na samples, permitting proper B. G. subtractions over the entire energy region to be made for these Na samples and for "open beam."

We find only the seven prominent resonances shown in Figs. 3(a)–3(c). Some weak levels observed in an earlier Nevis measurement by Garg *et al.*[20] were due to Fe resonances, since their samples were packaged in Fe-walled containers. A spectroscopic analysis of our Na sample material indicated that negligible impurities were present, with 0.003% Ca and 0.001% Al as the major impurities. The resonances which we report here are too strong to be due to impurities in the samples.

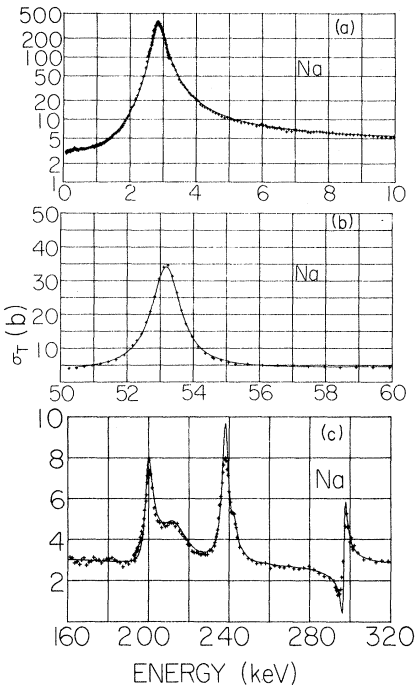

FIG. 3. The total cross section of ^{23}Na in energy regions where the cross section shows resonance structure. The experimental values are given by (+) and are derived from samples having $1/n = 4.8$ to 550 b/atom. Each experimental point represents a multichannel average of the data between 1 and 20 channels depending on the resolution and the rate at which σ_t varies. Typically, 5 channel averages were used in the high-energy region $E > 200$ keV between resonances, and 20 channel averages were used for $E < 50$ keV. The measured cross section may differ from the true cross section in regions where resolution and sample thickness effects are important. The solid lines shown in the figure parts (a), (b), and (c) are the result of R-matrix fits to the experimental data. The fit parameters are listed in Table II.

The curves shown in Figs. 3(a), 3(b), 3(c) are separate R-matrix fits to the total cross section over the energy intervals 0–10 keV, 50–60 keV, and 160–320 keV. The measured cross sections in the 10–50-keV and the 60–160-keV regions not shown here, were smoothly varying and showed no unambiguous resonance structure which could not be attributed to statistical fluctuations. The R-matrix fits in Figs. 3(a) and 3(b) each have only one Na level. The fit in Fig. 3(c) uses a five-level fit. Since natural Na is entirely ^{23}Na, with $I = \frac{3}{2}+$, s neutrons form states having $J = 1$ or 2(+), and $l = 1$ neutrons form states having $J = 0$, 1, 2, or 3(−). The neutron widths, Γ_n, for these levels are all large compared with their Doppler widths and with their capture widths, Γ_γ. Hockenbury *et al.*[16] find that $\Gamma_\gamma \approx 1$ eV for Na, so $\Gamma_n \approx \Gamma$ for these levels. The peak resonant cross section due to the (J, π) resonant part of the interaction is thus $g_J(4\pi/k^2) = (2830/E)\, g_J b$ (for E in keV). The spin weight factor $g_J = (2J+1)/2(2I+1)$ is $\frac{1}{8}$, $\frac{3}{8}$, $\frac{5}{8}$, and $\frac{7}{8}$, respectively, for $J = 0$, 1, 2, and 3. These quite different possible peak cross sections must be increased by $\lesssim 3$ b due to potential scattering contributions from the $l = 0$ parts of the neutron nucleus interaction not associated with the given level, and by the wing contribution from overlapping levels of different (J, π) where present. These possible σ values are well defined and quite different for the different allowed J value. For the completely resolved levels at 2.85 and 53 keV, the measurements establish level J values of 1 and 2, respectively. The directly evaluated thinnest sample, $1/n = 550$ b/atom, peak cross section for the 2.8 5-keV level was 410 b vs allowed peak resonance values of 124, 372, 621, or 870 b, respectively, for $J = 0$, 1, 2, or 3.

While it would, of course, have been very satisfying if the measured σ_{max} agreed more exactly with one of these "allowed" values, the residual uncertainties in some of our corrections to the data to obtain the "measured" σ_{max} were such as to be compatible with the difference between ~375 and 410 b/atom, but *not* compatible with such a large difference as that between 410 b/atom and ~124, 621, or 870 b/atom. Since the only possible values for σ_{max} are $\lesssim 3$ b/atom larger than 124, 372, 621, or 870 b/atom, clearly only ~375 b/atom is consistent with our "measured" value of 410 b/atom when the small uncertainties in certain of our experimental corrections to the data are considered. Thus we conclude that $J = 1$ is the proper choice for this resonance. Since we

believe that our "measured" σ_{max} should equal 375 b/atom, within statistical uncertainties if all experimental correction factors had been made correctly, there is the question of whether or not we should apply a small renormalization correction to the σ values for the $1/n = 550$ b/atom cross sections to achieve better agreement between measured σ_{max} and 375 b/atom. Since the subsequent interest, once the choice $J=1$ is established, is in obtaining a best-fit choice for Γ, we have made a "compromise" correction renormalizing factor to the $1/n = 550$ b/atom σ values which makes the "measured" peak σ at 2.85 keV = 385 b/atom. No such corrections were made to any of the data for the other resonances. This correction makes the comparison of the "width" of the experimental σ vs E curve with possible theoretical choices less biased by a small over-all upward shift in the measured curve which would tend to favor the choice of an overly large value for Γ. The renormalization of the $1/n = 550$ b/atom σ values also causes them to agree better with those for the thicker samples in their regions of meaningful overlap.

The choice of $l=0$ or $l=1$ is partly related to the expected interference effects between potential and resonance scattering for the resonant J state. This is particularly evident for the last level at 297 keV, and more weakly for the level at 2.85 keV. The level at 199 keV, which we assign $l=0$, has its interference effects largely masked by the wings of the p level at 212.5 keV.

The indicated fitting curves were generated using the Wigner-Eisenbud R-matrix analysis,[21] as modified by Thomas[22] to treat the weak capture contribution. We also use a modified form that sets the $l=1$ level shift factor equal to zero, so the energies will be those observed. Specifically, if $\hbar k$ is the center-of-mass neutron momentum, which is $A/(A+1)$ times the neutron lab momentum, then

$$\sigma_t(E) = (2\pi/k^2)\sum_j g_J[1 - \text{real}U_{l,J,j}(E)],$$

where

$$U_{l,J,j}(E) = e^{-2i\phi_l}\left[\frac{1+(S_l+iP_l)R_{l,J}}{1-(S_l+iP_l)R_{l,J}}\right].$$

The shift factor $S_0 \equiv 0$ for $l=0$. We arbitrarily set $S_1 = 0$ for $l=1$ so the level energies will be the measured values. (Note that these S_0 and S_1 shift parameters should not be confused with the strength functions.) The R-matrix formalism has

$$R_{l,J} = R^0_{l,J}(E) + \sum_J \frac{\gamma^2_{l,J,j}}{(E_{J,j} - E - i\Gamma_{j\gamma}/2)}.$$

The sum is over the levels included in each region of simultaneous fit.

The term $R^0_{lJ}(E)$ is a potential-scattering correction and has an energy dependence from the wings of levels outside the fitting region. We choose $R^0_{1,J} = 0$ (p levels) and, for $l=0$

$$R^0_{0,J} = A_J + B_J(E - E_1),$$

where E_1 is near the center of the fitted energy interval.

The P_l factors are barrier-penetration factors. We use $P_0(E) = ka$ and $P_1(E) = (ka)^3/[1+(ka)^2]$ in our energy region for Na. Here a is the square-well nuclear radius chosen as $1.41A^{1/3}$ fm ≈ 4.01 fm. This gives $(ka)^2 = E/1404$ keV, for laboratory neutron energy E.

The neutron partial widths $\Gamma_{l,J,j}$ are defined as equal to $2P_l\gamma^2_{l,J,j}$. The phase terms φ_l become $\varphi_0 = ka$ and $\varphi_1 \approx (ka)^3/3$. The capture widths, $\Gamma_{k\gamma}$, are treated as small corrections in the denominator terms in the expression for $R_{l,J}$.

A Fortran program was written by Dr. Rahn for use with the SEL810B computer system in the Columbia University Nuclear Engineering Department. The computer oscilloscope display gave a plot of the experimental σ vs E data points and the R-matrix fit for any chosen set of parameters E_1, A_1, B_1, A_2, B_2 and the J, l, E values for each resonance in the region. The data points and the fit curves could also be plotted using the computer attached Calcomp plotter, which feature was used to preserve and compare the displays for various "good fit" choices of parameters. An attempt to have the computer make a least-squares search for best-fit parameters was considered less desirable than using the physicist's judgment for the successive parameter changes, because of the need to vary several parameters simultaneously to remain in a good fit region, and also because the experimenter has better judgment

TABLE II. R-matrix parameter choices for the fits to the ^{23}Na levels shown in Fig. 3. For 0–10 keV, $A_1 = -0.260$, $A_2 = -0.575$, $B_1 = 0$, $B_2 = 0.005$, and $E_1 = 5$ keV. For 50–60 keV, $A_1 = -0.275$, $A_2 = -0.500$, $B_1 = -0.07$, $B_2 = 0$, and $E_1 = 55$ keV. For 160–320 keV, $A_1 = 0.45$, $A_2 = -0.50$, $B_1 = 0$, $B_2 = 0.0009$, and $E_1 = 300$ keV.

E (keV)	Γ (keV)	J	l
2.850	0.400	1	0
53.15	1.08	2	1
199.0	4.5	1	0
212.5	15	0	1
238	3.9	2	1
242.5	3.5	0	1
297.0	1.8	2	0

for regions where resolution effects are not negligible. Several many-hour sessions led to the parameter fit choices shown by the curves and by the parameters in Table II. These fits are not uniquely established, since the fitted curve changes only slowly when nearly compensating changes of more than one parameter are made together. The fits do not include resolution effects. They are sensitive to small changes in any of the continuous variables made one at a time. Comments on the fits, using the parameters in Table II are given below. Note that the Γ values are for the resonance energy. The fits include the energy dependence of each Γ away from exact resonance.

2.850 keV. As discussed above, the measured peak cross section is only compatible with $J=1$. We need $l=0$ to match the wing asymmetry. Our choice, $\Gamma=400$ eV is larger than the earlier, less exact Nevis evaluation of Garg *et al.*[20] for older data ($\Gamma=380$ eV, $J=2$, $l=0$). Our parameters are in good agreement with those of Moxon and Pattenden[23] and Lynn *et al.*[24] (Harwell).

53.15 keV. Our fit is excellent for $J=2$ and would be very poor for different J. A choice $l=0$ would yield a very asymmetric curve which could not fit the data. Various previously reported parameter fits by other groups, based on lower precision and poorer resolution measurements, have favored each of the choices $J=1$, 2, and 3. Our results are in general agreement with those of Moxon and Pattenden[23] (Harwell) $J=2$, $l=1$, $g\Gamma_n=750$ eV.

199.0 keV. Our fitting required that we use $l=0$, $J=1$. The only previous evaluations are from studies by Stelson and Preston[25] [Massachusetts Institute of Technology (MIT)] with assignments $J=1$, $l=1$, $\Gamma=5\pm2$, $E=204$ keV.

212.5 keV. This fit requires $J=0$, and thus $l=1$, for the peak cross section and for the general wing behavior. The older MIT evaluation[25] gave $E=217$ keV, $J=0$, $l=1$, $\Gamma_n=(14\pm10)$ keV.

238 keV. The resolution-limited measured peak cross section is too small for $J=3$ (13.3 b) and too large for $J=1$ (7.4 b) and seems best fitted by $J=2$, with $l=1$ required by the over-all fit wing behavior. The fit must include contributions from the poorly resolved 242.5-keV level which must have $J=0$, and thus $l=1$. The two resonances were treated as one in the MIT studies,[25] with $E=243$ keV, $J=1$ or 2, $l=1$, $\Gamma_n=(7\pm2)$ keV.

242 keV. See above.

297 keV. This level is seen with higher resolution in the Karlsruhe studies of Nebe and Kirouac.[15] They obtain $l=0$, $J=2$, $g\Gamma_n=1.18$ keV which agrees well with our assignments. They list the energy as 298.4 keV.

In addition to these levels, a number of small p levels seen by Hockenbury *et al.*[16] (RPI), Moxon and Pattenden[23] (Harwell), and Ribon *et al.*[26] (Saclay) in capture data at 7.5, 35.4, 114.7, 129.5, and 139.1 keV were not observed in our total cross-section measurements due to the narrow total widths of these levels. The levels which we studied all have $\Gamma_\gamma \ll \Gamma_n$ so that capture effects on our data analysis were negligible.

In view of the small number of levels treated, it is difficult to give very statistically significant evaluations of the s and p strength functions, S_0 and S_1, even when averaged over possible J values for each l. If the evaluations are made according to the rules obtained by Liou and Rainwater,[27] we obtain $10^4 S_0 = 0.24(1^{+1.91}_{-0.58})$. The uncertainty in S_1 is influenced by how many extra weaker p levels are believed actually to be present in the interval. For just the four p levels, $10^4 S_1 = 1.9(1^{+1.39}_{-0.51})$. If we assume that there are four extra p levels which give negligible contributions to S_1, but influence the sample size, then $10^4 S_1 = 2.06(1^{+0.77}_{-0.39})$. It should be noted that these evaluations are quite sensitive to the corrections of our assignments of levels as s or p wave. The above limits correspond to the 0.159 and 0.841 confidence limits, as explained in Ref. 27.

The results presented in this paper span an important energy region in the total cross section of sodium. Our measurements confirm the spin and parity assignment of $J=1$, $l=0$ for the important 2.85-keV level. In the energy region 100 to 300 keV, our data have higher resolution than the previous data set of Whalen and Smith[14] at ANL, and our observed σ_t indicates that the level at 238 keV is probably a doublet, with a peak cross section of >8.3 b. This peak cross section is in agreement with the data of Whalen and Smith. Above 280 keV, there is good agreement between our data and the Karlsruhe data set in places where the cross section is not rapidly varying. In places where the data sets overlap, the Karlsruhe data should be the preferred set because of their much higher resolution.

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