Neutron resonance spectroscopy: Chlorine*

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(Received 13 June 1974)

Results are presented of time of flight total σ vs E measurements for natural Cl to 400 keV. Resonance parameters were obtained for 35 resonances below 200 keV. Of these, 12 resonances were assigned to ^{35}Cl, 6 to ^{37}Cl, with 17 weak resonances for which isotope assignments could not be made. On the basis of our assignments as s or p resonances, $10^4 S_0 = (0.12^{+0.23}_{-0.07})$ for ^{35}Cl and $(0.59^{+0.66}_{-0.28})$ for ^{37}Cl. For ^{35}Cl, $10^4 S_1 = (0.77^{+0.53}_{-0.29})$ with no S_1 determination for ^{37}Cl. The anomalous behavior of σ vs E below $\sim$1000 eV for natural Cl, along with previously established thermal cross sections, are fitted by assigning a bound resonance in ^{35}Cl at -130 eV, having $l = 0$, $J = 2$, $\Gamma_n^0 = 0.725$ eV, and $\Gamma_\gamma = 0.378$ eV.

[NUCLEAR REACTIONS ^{35,37}Cl(n, n), (n, γ), $E = 20$ eV–400 keV; measured $\sigma_t(E)$; deduced E_0, $ag\Gamma_n$, l, J, S_0, S_1.]

I. INTRODUCTION

This paper presents the results of high resolution total cross section time of flight spectroscopy measurements for natural chlorine using the Columbia University Nevis synchrocyclotron. Samples of high purity CCl_4 were used, with the known smooth σ vs E for carbon subtracted to obtain curves of σ_t vs E to 400 keV for natural Cl. Resonance parameter assignments were obtained for 35 resonance to 200 keV. This is one of a series[1–9] of papers.

Natural Cl is 75.77% ^{35}Cl and 24.23% (at.%) ^{37}Cl, both having $I = \frac{3}{2}(+)$, with binding energies of 8.58 and 6.11 MeV, respectively, for an extra neutron. This is at a region of a minima of the s strength function, but is near a maxima of the p strength function. The number of resonances which we can assign to ^{35}Cl and ^{37}Cl, 12 and 6, respectively, are too small to yield a good statistical accuracy for the strength functions.

Previously reported curves of σ vs E for Cl, mainly from the pre-1960 studies of Brugger *et al.* (MTR),[10] Kiehn, Goodman, and Hansen (MIT),[11] and Newson *et al.* (Duke),[12] are given in the 1958 edition[13] of BNL-325. The results for earlier Columbia-Nevis Cl measurements of σ vs E were reported by Garg *et al.*[14]

The assignment of some Cl resonances to ^{35}Cl or ^{37}Cl using separated isotopes was made by Toller, Patterson, and Newson (Duke)[15] for a ^{35}Cl level at 26.5 keV, and for six levels to 94 keV in ^{37}Cl. Unfortunately, these assigned resonances are where level clusters are seen using higher resolution, so they cannot be used. The 0.398 keV resonance was assigned to ^{35}Cl by Brugger *et al.*[10] and by Popov and Shapiro.[16] The latter group[16] observed resonances in the Cl(n, p) reaction at 0.398 and 4.25 keV which is only possible for ^{35}Cl.

The main previous high resolution σ_t vs E studies for natural Cl, leading to resonance parameter assignments, are those of Patterson and Newson (Duke)[17] and of Morgenstern *et al.* (Saclay).[18] The measurements which we report here use comparable resolution to the Saclay[18] measurements, but extend to higher and lower energies. Our resonance parameters are mainly, but not always, in reasonable agreement with the Saclay[18] assignments. An earlier R-matrix analysis of this data was made by Singh.[19,20] The results presented in this paper are based on a complete more-thorough reanalysis of the data, starting with a reevaluation of background corrections etc., and using our usual "single level shape" and "area" analyses, which include the effect of Doppler broadening for narrow resonances. The present results are significantly different and supersede the earlier analysis results.

II. EXPERIMENTAL DETAILS AND PRELIMINARY DATA ANALYSIS

The time of flight transmission measurements used our 202.05 m flight path[1,3,7] with the cyclotron operating at 70 bursts/sec. Three sample thicknesses of pure CCl_4 were used having $(1/n) = 5.05$, 32.6, and 130.9 b/atom (of Cl), respectively. The entire spectrum for $E > 15$ eV was taken simultaneously using 16 000 detection channels. The first 10 240 channels (to 1279 eV) used 40 ns widths, fol-

lowed by 80 ns widths to 887 eV, and 160, 320, 640, and 1280 ns widths to 498, 221, 79.5, and 15 eV, respectively. About two hours counting time gave ~33×10^6 total histrogram counts for the thickest sample. About one hour each was devoted to counting with the two thinner samples in the beam, with ~25×10^6 histogram counts for each. To aid in evaluating the proper energy and transmission-sample-dependent background (BG) corrections, we also made "standard filter" measurements in which an Fe, Co, or Cu "standard" filter was kept in the beam, with and without the thick CCl_4 sample also in the beam. The standard filter method has been described in an earlier paper.[6] The idea is that the 7.6 cm Fe or 1.3 cm Co or Cu "filters" show strong "bottoming" ($T\approx0$) transmission dips at their strong s levels, recovering to higher count rate on the wings of the resonance, with the difference, wing to center, rates proportional to the filter level wing transmission. When the thick CCl_4 is also present, the wing to center count rate differences are reduced by the factor of the CCl_4 transmission in the (Fe, Co, or Cu) resonance wing region. This gives values for the CCl_4 transmission and, after subtracting the known σ vs E for carbon, σ for Cl, at these "calibration" energies. The subsequent analysis establishes the implied energy and sample dependent background values to be used for all three CCl_4 measurements, without the filters, at these energies and, by interpolation, between these energies.

Above 100 keV the background evaluation was less certain so we selected smooth corrections which yielded between level $\sigma(E)$ in agreement with the published values of Kiehn *et al.* (MIT).[11] The MIT measurements used monoenergetic neutrons and their experiment was less subject to uncertainties in the background correction. Since their measurements had much poorer energy resolution, only between level cross sections were compared. Below ~100 eV, there were no strong Cl resonances suitable for BG evaluation so the extrapolation from the BG correction near the 398 eV level was not good. We have thus made smooth energy dependent subtractions that produce reasonable fits to the $\sigma(E)$ results of Brugger *et al.*[10] near 20 and 100 eV.

The observed resonances were classified as having $l=0$ only if a shape asymmetry, due to interference between resonance and potential scattering, was observed. The levels classed as $l=1$, lacked such asymmetry. It was established that all of our resonances were due to Cl by two methods. A spectroscopic analysis was made by Lucius Pitkin, Inc. of a sample taken from the CCl_4 supply from which all of the samples were prepared. It showed no significant abundance of any of 33 elements for which tests were made. The main contaminants of *a priori* concern involved halogen atoms (Br, I) substituting for Cl atoms. A careful study of the data at energies of the strongest Br or I resonances revealed no trace of these elements.

Since there are four possible s level populations, and eight p level populations, level repulsion effects are expected to be unimportant. There were, in fact, level clusterings in certain energy regions such as at (14.8, 16.3, and 17.1 keV), and at (25.6, 26.6, 27.3, and 27.8 keV), while no levels could be seen in other extended regions such as those from 18 to 25 keV and 70 to 90 keV. One would expect the members of each cluster to have at most one level from each of the 12 $(s+p)$ populations due to level repulsion effects within each population.

In the absence of Doppler broadening, the peak cross section at resonance due to the state involved is $2.6\times10^6(1\text{ eV}/E)[(A+1)/A]^2 ag\Gamma_n/\Gamma$ b/natural Cl atom. Here a is the isotope fractional abundance and $g=(2J+1)/2(2I+1)$ is the spin statistical weight. For ^{35}Cl, the factor $ag=0.663$, 0.474, 0.284, and 0.0947, respectively, for $J=3,2,1,0$ with the corresponding values for ^{37}Cl smaller by a factor 0.320. For those resonances where we give independent A assignments, it is on the basis of the peak resonance cross sections and the differences in the predictions for $A=35$ or 37. This usually permits both l and J to be assigned for the resonance. Since there are four s populations, and since the potential scattering cross section is small, the level asymmetry for s levels is not large. A few levels have less certain $l=0$ assignments. In all cases, we give values of $ag\Gamma_n$ for the resonance.

III. RESULTS AND DISCUSSION

Above ~2 keV, the potential scattering cross section for Cl is found to be $\lesssim1.5$ b, but there is a gradual increase of over a factor of 10 in σ_t below 1 keV neglecting the resonance structure near 398 eV. This behavior is attributed to a negative energy (virtual) s level in ^{35}Cl. Previous evaluations have assigned E_0, Γ_n^0, and Γ_γ values for this level to best fit the thermal neutron (n,γ), (n,p), and scattering cross sections for natural Cl, and the measured shape of the drop-off of σ vs E below ~1 keV. The values of E_0, Γ_n^0, and Γ_γ are coupled in each choice to fit the thermal neutron cross sections. Previously reported values are summarized in BNL-325 (1964).[17] Values of $(-E_0)$ of 0.210 ± 0.010, 0.140 ± 0.005, and 0.075 keV have been given for the favored choice $J=2$. These values differ by amounts large compared with their quoted uncertainties.

Figure 1 shows a fit to our measured σ vs E below 3 keV by a single virtual Breit-Wigner level using $E_0 = -130$ eV, $l = 0$, $J = 2$, $\Gamma_n^0 = 0.725$ eV, $\Gamma_\gamma = 0.378$ eV, and $R' = 3.33$ fm ($4\pi R'^2 = 1.39$ b/atom) for each of the four $l = 0$ spin-isotope states of natural Cl ($J = 1, 2$, $A = 35, 37$). The known thermal cross sections for capture, coherent scattering, and total scattering give additional information which, with our σ vs E of Fig. 1, require these values. We use the recommended[21] values for the $\sigma(n,\gamma)$ thermal cross sections of natural Cl, ^{35}Cl, and ^{37}Cl of (33.2 ± 0.5), (43 ± 2), and (0.433 ± 0.006) b/atom. These are weighted averages of measurements by many groups.[21] Let f_j, g_j, and a_j ($j = 1$ to 4) be, respectively, the isotopic abundance, the spin statistical weight, and the scattering length for each of the four $l = 0$ states of natural Cl. The coherent scattering length $a_{\text{coh}} = \sum f_j g_j a_j = (9.605 \pm 0.003)$ fm from measurements of Koester and Nistler[22] based on liquid mirror total reflection measurements. The recommended[13] value $\sigma = (16 \pm 3)$ b/atom for natural Cl thermal scattering is less precisely known, but is consistent with Fig. 1. The small (n, p) and (n, α) cross sections give negligible contributions. The thermal capture, if assigned to the virtual level, establishes $fg\Gamma_n^0\Gamma_\gamma E_0^{-2}$ for the resonant state. If $a_j = R' = 3.33$ fm except for the resonant state, with $a = (R' + a_r')$ for that state, $a_r' = \lambda_{1\,\text{eV}}\Gamma_n^0/2E_0$, this establishes $fg\Gamma_n^0/E_0$ for the resonant state. The choice $J = 1$ does not give a reasonable fit to the thermal scattering or to our σ vs E in Fig. 1. The Breit-Wigner resonance denominator, $(E - E_0)^2 \gg \Gamma^2$, for any choice of E_0, determines the shape of the drop-off of the fit curve in Fig. 1. The fit parameters in Fig. 1 agree with σ_γ and a_{coh}(thermal) and give thermal

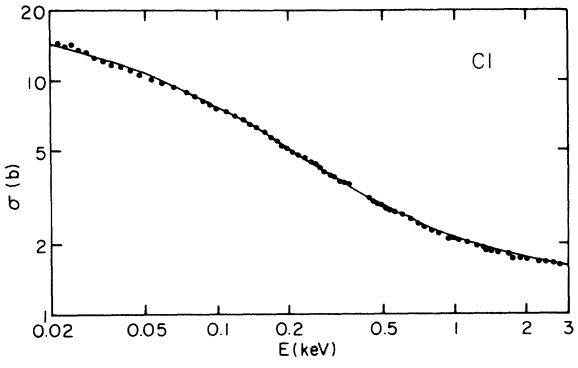

FIG. 1. A theoretical fit (solid curve) to the data (shown by circles) from ~20 eV to 3 keV using a bound level at $E_0 = -130$ eV belonging to ^{35}Cl with $l = 0$, $J = 2$, $\Gamma_n^0 = 0.725$ eV, $\Gamma_\gamma = 0.378$ eV, $R' = 3.33$ fm ($\sigma_p = 1.39$ b). The experimental thermal values of $\sigma(n, \gamma) = (33.2 \pm 0.5)$ b and $\sigma_s = (16 \pm 3)$ b are in excellent agreement with implied values of 33.13 and 16.68 b, respectively.

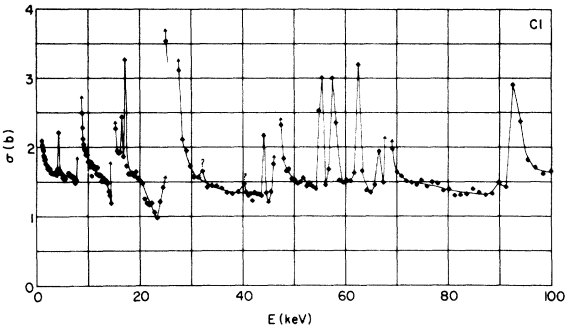

FIG. 2. The measured thick sample ($1/n = 5.05$ b/atom) σ_t vs E for natural chlorine from about 1 keV to 100 keV using many channel averages. The plot emphasizes the cross section behavior between levels, with no attempt to indicate peak cross section at resonances.

$\sigma_s = 16.68$ b/atom.

Figure 2 shows the measured σ vs E for Cl below 100 keV. The points are averages over many adjacent channels to reduce statistical fluctuations and emphasize between-resonance behavior, and resonance wings, with high σ points near resonance omitted. The results for the resonance parameter assignments are given in Table I for resonances to 200 keV. Figure 3 shows σ vs E from the thick sample data above 100 keV and shows the observed resonances without using multichannel averaging. Some of the weak structures, which could be either levels or statistical fluctuations, have question marks. An inspection of Table I

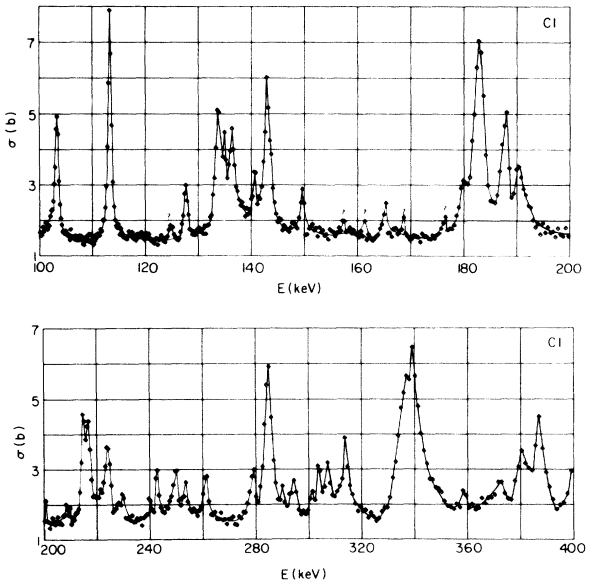

FIG. 3. The measured single channel σ_t vs E for the thickest Cl sample from 100 to 400 keV.

TABLE I. Neutron resonance parameters for levels in natural Cl below 200 keV. Letters a, b, c after the isotope assignment are from Refs. 10, 16, and 15, respectively, where separated isotopes were used (10 and 15) or the (n,p) reaction (^{35}Cl) resonance was observed.

E_0 (keV)	A	l	J	$ag\Gamma_n$ (eV)
0.3977 ± 0.0004	35 [a,b]	1		0.028 ± 0.003
4.2481 ± 0.0015	35 [b]	1		0.14 ± 0.02
8.3121 ± 0.0083	37 [c]	0	2	11.5 ± 1.0
14.788 ± 0.010	35	0	2	14.0 ± 2.0
16.340 ± 0.012		1		2.65 ± 0.30
17.120 ± 0.012	35	1	(2)	7.0 ± 1.0
25.563 ± 0.045	37	(0)	1	60.0 ± 10.0
26.601 ± 0.024	35	(0)	2	55.0 ± 10.0
27.316 ± 0.025	37	1	0	1.5 ± 0.5
27.805 ± 0.026		1		0.6 ± 0.3
44.12 ± 0.05		1		4.0 ± 1.0
46.58 ± 0.06	37 [c]	0	2	65.0 ± 10.0
51.53 ± 0.06		1		1.2 ± 0.4
54.90 ± 0.07		1		14.0 ± 4.0
55.36 ± 0.07		1		18.0 ± 4.0
57.72 ± 0.08	35	1	1, 2	46.0 ± 5.0
62.67 ± 0.09		1		35.0 ± 5.0
66.74 ± 0.10		1		8.0 ± 2.0
68.18 ± 0.10	37 [c]	0	2	55.0 ± 5.0
90.25 ± 0.15		1		6.0 ± 3.0
93.06 ± 0.16	37 [c]	0	2	98.0 ± 8.0
103.3 ± 0.2	35	1		100.0 ± 20.0
113.3 ± 0.2	35	0	2	220.0 ± 30.0
127.5 ± 0.3				52.0 ± 8.0
133.9 ± 0.3				200.0 ± 50.0
135.0 ± 0.3				50.0 ± 10.0
136.3 ± 0.3				150.0 ± 40.0
140.7 ± 0.3				45.0 ± 10.0
142.9 ± 0.3	35	1		360.0 ± 50.0
149.6 ± 0.3				50.0 ± 10.0
165.3 ± 0.4				48.0 ± 10.0
180.0 ± 0.4				150.0 ± 50.0
182.8 ± 0.5	35	1		900.0 ± 200.0
188.1 ± 0.5	35		1	450.0 ± 50.0
190.1 ± 0.5	35		1	300.0 ± 40.0

shows that 12 resonances were assigned to ^{35}Cl, six to ^{37}Cl, and 17 weak resonances could not be assigned A values.

From the results for the resonance parameter assignments in Table I, we can obtain implied values for the s and p strength functions, averaged over J. For ^{35}Cl, there are three $l=0$, $J=2$ resonances at 14.8, 26.6, and 113.3 keV. This implies that $10^4S_0 = (0.12^{+0.23}_{-0.07})$. There are five levels assigned to ^{35}Cl, $l=1$ below 105 keV. In addition, there are nine levels classified as $l=1$ in this energy interval for which no isotope assignment has been made. If, for these later resonances, half of their $\sum g\Gamma_n^1$ is added to that for ^{35}Cl, we obtain $10^4S_1 = (0.77^{+0.53}_{-0.29})$ for ^{35}Cl. We have five resonances between those at 8.3 and 93.1 keV which are considered to be s levels in ^{37}Cl. This gives $10^4S_0 = (0.59^{+0.66}_{-0.28})$ for ^{37}Cl. Four of the levels are assigned to be $l=0$, $J=2$ (^{37}Cl). This gives $10^4S_0 = (0.65^{+0.90}_{-0.33})$ for $J=2$ levels only in ^{37}Cl. The above uncertainties are assigned by the methods of Ref. 23. The main comparison is with the Saclay results. They list their E_0, A, $g\Gamma_n$, l, and J for levels from 14.7 to 113 keV. Even where they do not assign A, they give $g\Gamma_n$ rather than $ag\Gamma_n$, without explanation. This makes comparison difficult. When their $g\Gamma$ values are converted to $ag\Gamma_n$ values where they give A and $g\Gamma_n$, we agree on $ag\Gamma_n$ much better than our combined uncertainties would suggest. There are some levels where they give A but we do not. Our l assignments differ from four of theirs. We assign the 68.18 keV level to ^{37}Cl on the basis of the work of Toller *et al.*,[15] while they choose $A=35$. We differ on l assignment for the strong level at 113.3 keV. Our $l=0$ strength functions agree with theirs. Our energies are slightly different. It is harder to compare with earlier, poorer resolution measurements where not all levels were resolved and the energy equivalences may be ambiguous. The agreement is fair for levels below 14 keV.

We wish to thank Dr. S. Wynchank and Dr. H. S. Camarda for their participation in the experiment. The technical assistance provided by W. T. Marshall and C. Gillman was important.

*Research supported in part by the U. S. Atomic Energy Commission.

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