M/L- and L/K-Orbital Electron-Capture Ratios and Transition Energy in Cd^{109} Decay*

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A direct measurement of the M/L- and L/K-orbital electron-capture ratios in the decay of Cd^{109} was carried out by using a gaseous radioactive source of $Cd^{109}(CH_3)_2$ in a multiwire proportional counter with internal anticoincidence. The experiments yielded the following values of the orbital electron-capture probability ratios: $P_M/P_L=0.232\pm0.020$, $P_{L+M+\ldots}/P_K=0.332\pm0.015$, and $P_L/P_K=0.237\pm0.015$. From the P_L/P_K value, the transition energy for capture to the 88-keV ($\frac{7}{2}+$) state in Ag^{109} was found to be $Q_{EC}=80_{-3}^{+7}$ keV by application of the theory. The experimental M/L-capture ratio is in excellent agreement with the exchange-corrected theoretical result extrapolated from Bahcall (0.230) and with the theoretical results of Robinson.

I. INTRODUCTION

THE now well-established discrepancy between precision experimental values of the L/K-capture ratio in allowed radioactive decay and the theoretical values of Brysk and Rose[1] was first noted by Robinson and Fink.[2] Odiot and Daudel[3] calculated electron exchange effects for Ar^{37} decay and found that these effects increased the theoretical L/K-capture ratio by about 20%, bringing the theory into good agreement with later precision experiments. A detailed general treatment of these exchange-overlap corrections was given by Bahcall[4] in the region of $13\leqslant Z\leqslant 37$, where analytic Hartree-Fock wave functions are available from the work of Watson[5] and Watson and Freeman.[5] A comparison of the recent precision experimental values of L/K-capture ratios, from multiwire proportional counter measurements on internal gaseous sources, has been made by Moler and Fink[6] and summarized by Bahcall.[4]

Bahcall[4] found that the atomic overlap effect reduces all capture rates slightly and that electron exchange reduces the K-capture rate while enhancing L_I- and M_I-capture rates, the M_I-capture rate being increased more than the L_I rate. These corrections all are decreasing functions of the atomic number.

The success of the exchange-overlap corrections for L/K capture for allowed decay led Bahcall to extend the theoretical calculations for allowed electron capture to the M shell[7] in the region $13\leqslant Z\leqslant 37$. The theoretical M/L-electron capture probability ratio is, according to Bahcall,[7]

$$P_M/P_L=(P_{M_I}/P_{L_I})^0[X^{M/L}+M_{II}/M_I-L_{II}/L_I], \quad (1)$$

where $(P_{M_I}/P_{L_I})^0$ is the usual ratio of M_I/L_I capture, calculated according to the method of Brysk and Rose[1]; $X^{M/L}$ is the exchange-overlap correction to the usual M_I/L_I capture ratio; and M_{II}/M_I and L_{II}/L_I are the respective ratios of capture of $P_{1/2}$ electrons to s electrons in the M and L shells. The terms M_{II}/M_I and L_{II}/L_I almost exactly cancel in Eq. (1). Numerical values of the correction factor $X^{M/L}$ and of the usual ratio of $(P_M/P_L)^0$ for s electrons are given by Bahcall[7] enabling the calculation of theoretical M/L-capture ratios that are accurate to within 2%. Values of $X^{M/L}$ in the region $13\leqslant Z\leqslant 37$ are represented to within 0.5% by the formula[7]

$$X^{M/L}=1+5.593/Z-59.5/Z^2+1111/Z^3. \quad (2)$$

Relativistic effects on P_M/P_L are of the order of $(\alpha Z)^2$ and hence are small.[8] Nuclear-size effects amount to less than 0.3% for[9] $Z\leqslant 50$[9] and thus are negligible.

Hubbard[10] has calculated the theoretical M-shell contribution to orbital-electron capture by the method of Brysk and Rose,[1] but without overlap-exchange corrections, in the region $60\leqslant Z\leqslant 90$. In this work the calculations of L- and K-capture rates were extended to third-forbidden and third-forbidden-unique transitions and indicate that, at least for unique-forbidden cases, the M/L-capture ratio exceeds 20% for all values of the transition energy in this region of $Z\geqslant 60$. If both the L/K- and the M/L-capture ratios are known for nonunique forbidden decays, one has the possibility of calculating the absolute value of the nuclear matrix element.[10]

Robinson[11] has computed allowed M_I/L_I wave func-

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[1] H. Brysk and M. E. Rose, U. S. Atomic Energy Commission Report, ORNL-1830, 1955 (unpublished); Rev. Mod. Phys. **30**, 1169 (1958).
[2] B. L. Robinson and R. W. Fink, Rev. Mod. Phys. **27**, 424 (1955); **32**, 117 (1960).
[3] S. Odiot and R. Daudel, J. Phys. Radium **17**, 60 (1956).
[4] J. N. Bahcall, Phys. Rev. Letters **9**, 500 (1962); Phys. Rev. **129**, 2683 (1963); **132**, 362 (1963).
[5] R. E. Watson, Phys. Rev. **118**, 1036 (1960). R. E. Watson and A. J. Freeman, *ibid.* **123**, 521 (1961); **124**, 1117 (1961).
[6] R. B. Moler and R. W. Fink, Phys. Rev. **131**, 821 (1963).
[7] J. N. Bahcall, Phys. Rev. **131**, 1756 (1963).
[8] D. Layzer and J. N. Bahcall, Ann. Phys. (N. Y.) **17**, 177 (1962).
[9] I. M. Band, L. N. Zyrianova, and T. Chen-Zhui, Izv. Akad. Nauk SSSR, Ser. Fiz. **20**, 1389 (1956).
[10] W. M. Hubbard, Phys. Rev. **137**, B245 (1965).
[11] B. L. Robinson, Nucl. Phys. **64**, 197 (1965).

tion ratios for the region $55 \leqslant Z \leqslant 90$, using the relativistic Thomas-Fermi-Dirac-screened wave functions for finite-size nuclei given by Brysk and Rose[1] and by Brewer, Harmer, and Hay.[12] Robinson's values, together with the allowed M/L-capture ratios calculated by Bahcall[7] with correction for overlap exchange, are plotted in Fig. 1 as a function of Z. An extrapolation of Bahcall's corrections[7] suggests that the allowed M_I/L_I-capture ratio may be about 9% larger than the wave function ratio at $Z=55$ and about 6% larger for $Z=90$. The application of these corrections places the theoretical M_I/L_I-capture ratios of Robinson[11] on the dashed curve in Fig. 1,[11] which then connects smoothly to the more detailed results of Bahcall[7] for $Z \leqslant 37$.

The first experimental measurement of M capture was reported by Manduchi and Zannoni,[13] who found $P_M/P_L=0.141\pm0.010$ for Ge^{71} decay, using gaseous $Ge^{71}H_4$ in a multiwire proportional counter. The wave functions of Watson-Freeman[5] and of Hartree, Hartree, and Manning[14] without exchange-overlap corrections both give a theoretical value of the M/L-capture ratio for Ge^{71} decay of 0.149. The exchange-corrected value calculated by Bahcall[7] is 0.173, in serious disagreement with the experimental value. However, the M-shell binding energy in germanium is only some 160 eV, which results in only about 6 primary ion pairs. Such a small number of ion pairs renders the determination of the intensity of the M peak extremely difficult and rather susceptible to losses due to electron attachment by impurities in the counting gas, particularly at the high pressure (6 atm) used.[13] Such losses in the Ge^{71} experiment would affect the M peak more than the L peak and would result in a low value for the experimental M/L-capture ratio. In the case of Cd^{109}, the M-shell binding energy is approximately 719 eV which is sufficiently high to avoid this difficulty.

It is clear that additional precision M/L-capture ratio measurements for allowed transitions are of interest in testing the theory and especially the exchange-overlap corrections. For this reason, we have investigated the capture ratios of Cd^{109} reported here, as part of a continuing study.

II. EXPERIMENTAL

The multiwire proportional counter with internal anticoincidence used in the present work was described previously.[15] The counter can be operated at pressures up to at least 5 atm. The preamplifier[16] output from

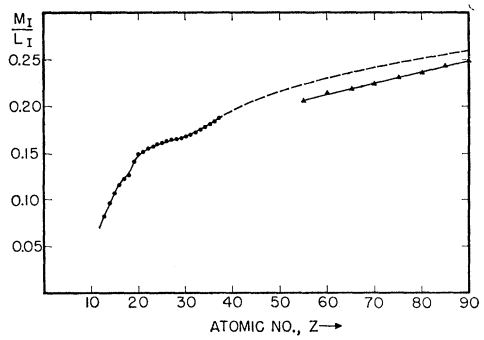

FIG. 1. Allowed M_I/L_I capture and wave function ratios evaluated at the nuclear surface for finite-sized nuclei, omitting energy dependence (see Ref. 11). The points in the region $13 \leqslant Z \leqslant 37$ are from Bahcall (Ref. 7) and include the exchange-overlap corrections in detail. The points for $55 \leqslant Z \leqslant 90$ are Robinson's values (Ref. 11) of screened relativistic wave function ratios for finite-sized nuclei: if Bahcall's overlap-exchange corrections are extrapolated into this region of atomic number, the allowed M_I/L_I-capture ratios fall on the dashed curve.

the central counter was fed to the amplifier input of an RIDL 200-channel transistorized analyzer. The gain of the anticoincidence ring counter could be adjusted through the use of an auxiliary separate 0- to 500-V negative power supply having stability of 0.001%. Pulses from the ring counter, after amplification, were used to control a linear anticoincidence gate, the length of which could be varied from 10 to 200 μsec; in the present work, the length was 100 μsec. These anticoincidence pulses were fed to the blocking input of the analyzer. Pulses at this input also were coupled internally to the analyzer's live timer to correct for deadtime caused by high counting rates in the ring counter. The bias was set at 10 keV for the ring counter since lower bias settings had no effect on the results, but increased the deadtime considerably.

The source was in the form of gaseous radioactive $Cd^{109}(CH_3)_2$, which is liquid at room temperature and boils at 105°C, synthesized[17] by a Grignard reaction on anhydrous radioactive $CdCl_2$:

$$Cd^{109}Cl_2+2CH_3MgI \xrightarrow{Et_2O} Cd^{109}(CH_3)_2+MgI_2+MgCl_2.$$

The anhydrous $CdCl_2$ was prepared by refluxing radioactive $CdCl_2$, with thionyl chloride, $SOCl_2$, under an argon atmosphere. After drying the $CdCl_2$ under vacuum, the Grignard reagent was added. This reagent was prepared by treatment of Mg turnings in anhydrous diethyl ether with CH_3I. A small quantity of I_2 was added to initiate the reaction, which commenced on adding the CH_3I dropwise to the warmed mixture. The reaction forming $Cd(CH_3)_2$ goes to completion rapidly, so that stirring was discontinued after about one hour. Since $Cd(CH_3)_2$ boils at 105°C, it was purified by

[12] H. R. Brewer, D. S. Harmer, and D. H. Hay, Phys. Rev. Letters **7**, 319 (1961).
[13] C. Manduchi and G. Zannoni, Nucl. Phys. **36**, 497 (1962); Nuovo Cimento **24**, 181 (1962).
[14] W. Hartree, D. R. Hartree, and M. F. Manning, Phys. Rev. **59**, 299, 306 (1941).
[15] J. Scobie, R. B. Moler, and R. W. Fink, Phys. Rev. **116**, 657 (1959).
[16] Construction details of the counter together with details of the newer electronic circuitry used in the present investigation are summarized in the Ph.D. thesis of R. B. Moler, University of Arkansas, 1964 (unpublished).
[17] G. E. Coates, *Organometallic Compounds* (Methuen & Company, Ltd., London, 1960).

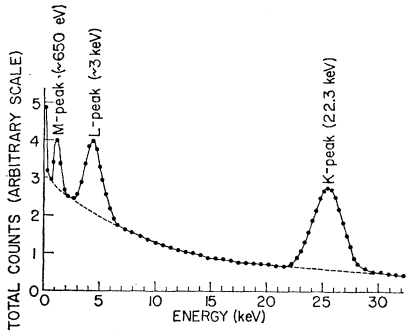

FIG. 2. The M, L, and K peaks from Cd^{109} decay in the central counter in anticoincidence with the ring counter. These peaks lie at energies of the order of 650 eV, 3 keV, and 22 keV, respectively, and the respective full width at half maximum (FWHM) resolutions are 50, 22, and 14% at 4-atm counter pressure. The background, which is not subtracted, appears as a dashed line and is considerably larger than the natural background of the counter when the source is absent. The high background arises from conversion electrons from the 88-keV gamma transition and from photoelectrons from the wall-effect caused by unconverted 88-keV gammas and escaping noncoincident K x rays. Since the 88-keV transition is delayed because of its 40-sec half-life in Ag^{109m}, its effects are not removed by the anticoincidence ring counter (see discussion in text).

fractional distillation. A first distillation was performed at atmospheric pressure, the ether collecting first in a dry ice-acetone trap until the temperature rose to 100°C, at which point a second collection trap was substituted. The collection was terminated at 125°C. A final fractional distillation was carried out under reduced pressure, the boiling point being 50°C, and the middle fraction (2 ml) of $Cd^{109}(CH_3)_2$ was collected for subsequent introduction into the counter.

The vacuum-filling system for the counter[16] enabled the sample to be recovered and reintroduced into the counter after evacuation. In all runs the partial pressure of radioactive $Cd(CH_3)_2$ was kept below 1.0 Torr, no more than 50 mg being introduced into the counter in any one run. The counter, of about 10-liter volume, was filled to 3, 4, or 5 atm through a purifier containing hot calcium (400°C) with a mixture of 10% methane–90% argon, and it was allowed to remain overnight before spectra were taken to insure proper mixing of source and counter gases. In a preliminary experiment, nonradioactive $Cd(CH_3)_2$ was introduced at a rather high pressure (10 Torr) along with some Fe^{55}-labeled ferrocene vapor as a standard. The resolution remained good at pressures up to 4 atm.

The first runs showed very little contamination of the central counter after evacuating and refilling. On very long exposure, however, of the order of days, some activity deposited on the walls and wires in the counter, but the amount never was more than a few percent of the total activity within the counter. Some runs at 2 and 3 atm were made at elevated temperature ($\approx$70°C). No change in the gross activity of the center counter was observed, indicating that none of the source had condensed on the walls. At the higher temperature a slight increase in gas multiplication was noted, but with no decrease in resolution. Nagantani and Sakaki[18] reported that a radial thermal gradient had a large effect on gas multiplication; however, in our case the thermal gradient was small, since the counter was heated uniformly from the outside and allowed to come to thermal equilibrium.

A typical spectrum showing M, L, and K peaks from Cd^{109} decay is shown in Fig. 2, which represents the central counter in anticoincidence with the ring counter. Figure 3 shows the K peak and the 88-keV gamma peak without anticoincidence. These spectra were obtained at a counter pressure of 4 atm. The background is very much larger than the natural background of the counter when the source is not present. The explanation of this background is to be found in the noncoincidence of the 88-keV gamma, shown in the decay scheme in Fig. 4. This gamma transition is approximately 90% converted.[19] There is some probability, additionally, of photoelectrons being produced at the walls and wires by the approximately 10% of

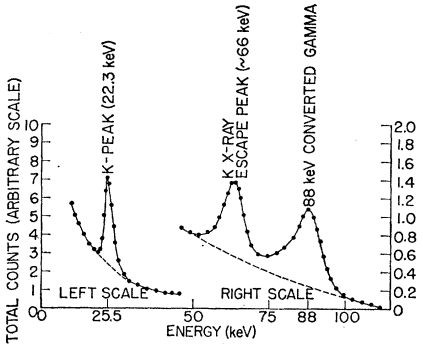

FIG. 3. Representative spectrum showing the K- and 88-keV gamma peaks (left- and right-hand scales, respectively) from the central counter without anticoincidence at 4 atm pressure. The background is shown as a dashed line and has not been subtracted. The FWHM resolution of the K peak is 14% and of the 88-keV gamma peak, 10%. The peak at about 66 keV is due to K x-ray escape following the 88-keV transition.

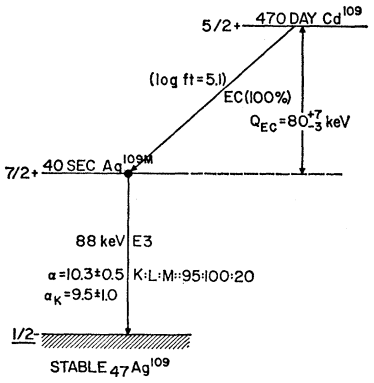

FIG. 4. Decay scheme of Cd^{109} (Ref. 19). Note that the 88-keV gamma transition is delayed owing to its 40-sec half-life and therefore is not coincident with the x ray and Auger transitions following electron capture in Cd^{109}. See discussion in text.

[18] T. Nagatani and Y. Sakaki, Rev. Sci. Instr. 33, 556 (1962).
[19] *Nuclear Data Cards* (National Research Council—National Academy of Sciences, Washington 25, D. C.).

88-keV gammas that remain unconverted, as well as by the escaping noncoincident 22-keV K x rays following conversion of the 88-keV transition. Since this transition in Ag^{109} is delayed due to its half-life of 40 sec, its effects are not coincident with the x-ray and Auger transitions immediately following orbital electron capture in Cd^{109}, and thus are not removed by the anticoincidence ring counter. (The 88-keV unconverted gammas have negligible interaction probability in the counter gas, the half-thickness in 4 atm of argon being greater than 10 m.) The high background was therefore interpolated beneath the M, L, and K peaks (Fig. 2) and subtracted in determining the relative areas of these peaks with a planimeter.

III. EVALUATION AND DISCUSSION OF RESULTS

A. The M/L-Capture Ratio

With a gaseous internal source in the multiwire counter the L and M x rays and Auger electrons will be completely stopped in the gas volume, while a fraction of the K x rays escapes. If a K_α x-ray escapes, the vacancy left in the $L_{II,III}$ subshells will appear as an L-capture event, increasing the L/K-capture ratio. To correct for these effects, a knowledge is required of the K fluorescence yield,[20] ω_K, and k_α and k_β, the fraction of K_α and K_β x rays, respectively, in the K series of x-ray transitions of the daughter ($Z=47$).[21]

In the present case, assuming that M and L x-ray escape are negligible, the corrected experimental M/L-capture ratio can be obtained in the following manner: The observed number of events in the M peak is

$$N_M=P_M+P_K\omega_K Pk_\beta, \tag{3}$$

and the observed number of events in the L peak is given by

$$N_L=P_L+P_K\omega_K Pk_\alpha, \tag{4}$$

where P_M and P_L are the respective number of M-capture and L-capture events occurring in the central counter volume, and P is the total probability of K x-ray escape from the central counter. The ratio of Eqs. (3) and (4) is

$$N_M/N_L=\frac{(P_M+P_K\omega_K Pk_\beta)}{(P_L+P_K\omega_K Pk_\alpha)}. \tag{5}$$

Solving Eq. (5) for the ratio P_M/P_L, we obtain

$$P_M/P_L=(N_M/N_L)[1+(P_K/P_L)\omega_K Pk_\alpha]-(P_K/P_L)\omega_K Pk_\beta. \tag{6}$$

In Eq. (6) the terms involving ω_K, k_α, k_β, and P have opposite signs, so that they partially cancel. As discussed below, this fact takes on great significance in measurement of the M/L-capture ratio. The P_L/P_K ratio may be written in the form

$$P_L/P_K=(N_L/N_K)(1-P\omega_K)-P\omega_K k_\alpha, \tag{7}$$

where N_K is the observed number of events in the K peak.

An improved method of computing the K x-ray escape probability P has been reported earlier by Moler and Fink,[6] but it is not valid in the present situation for the following reason. The previous method[6] of calculating P from reactor theory applies when the relaxation length λ, defined as $1/\mu_m p$, where μ_m is the mass attenuation coefficient and p is the density of the gas, is less than the counter diameter. For a 22.3-keV x ray passing through 4 atm of argon, $\lambda\approx 25$ cm, whereas the counter diameter is only 5 cm. Obviously the value of P will be large and cannot be evaluated by the previous method.[6]

Therefore, in order to evaluate P, we made direct experimental measurements at various pressures by using the following method. By measuring the ratio of the total number of conversion electrons from the 88-keV transition in Ag^{109} (Fig. 4) to the number of K-conversion electrons resulting in escaping K x rays, a direct determination is made of P_e, the fraction of K x rays which escape the central counter and which trigger the anticoincidence ring counter. The relative intensities of the K-, L-, and M-conversion electrons in Cd^{109} decay have been measured with magnetic beta spectrometry,[19] and these, together with the values of k_α, k_β,[21] and ω_K,[20] enable one to determine the value of P_e. In terms of the ratio of the intensity of the K x-ray escape peak N_e to the total intensity of the converted gamma transitions N_T (the ratio of the escape/gamma peak areas in Fig. 3), the K-escape probability P_e can be written

$$P_e=N_e/(N_T f_K\omega_K), \tag{8}$$

where f_K is the fraction of the total conversion occurring in the K shell.[19] From measurements of the relative intensity of the K-escape peak to the converted gamma peak N_e/N_T, Fig. 3, the value of P_e was determined for our experiment at various pressures from 1 to 5 atm. The values found are $P_e=1.0$ at 1 atm, 0.95 at 2 atm, 0.88 at 3 atm, 0.82 at 4 atm, and 0.75 at 5 atm

In addition to a knowledge of P_e, however, it is necessary to know the fraction of K x radiation which does not trigger the anticoincidence gate. A total escape probability P is then obtained for use in Eqs. (6) and (7). A way to measure this fraction of escaping K x rays which do not trigger the gate is to observe coincidences between the 88-keV converted gamma events recorded in the central counter and K x rays recorded in the ring counter. Knowing the fraction of K x rays arising in the central counter due to K con-

[20] H. Mark, R. C. Jopson, C. D. Swift, and R. W. Fink (unpublished).

[21] A. H. Wapstra, G. J. Nijgh, and R. van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).

TABLE I. Experimental determination of the P_M/P_L ratio in Cd^{109} decay.

Counter pressure (atm)	No. of runs	Total K x-ray escape probability P	Observed intensity ratio N_M/N_L	Experimental value P_M/P_L from Eq. (6)
3	8	0.65	0.207±0.015	0.234±0.026
4	4	0.50	0.205±0.017	0.225±0.029
5	1	0.40	0.211±0.025	0.233±0.033
			Average value	=0.232±0.020

version of the gamma transition,[19] the coincidence rate measures the fraction of the escaping K x rays stopped in the ring counter. This fraction is about 25% for a window set to include 20–30 keV. On the other hand, reducing the lower bias level to 15 keV increased the coincidence rate somewhat. When the window was adjusted to include 10–30 keV, the coincidence rate indicated that approximately 50% of escaping K x rays triggered the anticoincidence gate at 4-atm pressure. At 3 atm, this fraction decreased to 35%, becoming rapidly smaller with decreasing pressure and approaching 0% at 1 atm. The error involved in these results amounts to about 25%.

In Eqs. (6) and (7) for P_M/P_L and P_L/P_K, respectively, the factor of greatest indeterminancy is the total escape probability P, the large value of and uncertainty in which shows the importance of the fact that in Eq. (6) for a precision measurement of the M/L-capture ratio, the terms involving P largely cancel. In the present case, P_M/P_L varies only by about 20% as P is varied from 0 to 1. Thus, the M/L-capture ratio is very much less sensitive to uncertainties in ω_K, k_α, k_β, and P [Eq. (6)] than is the L/K-capture ratio [Eq. (7)]. The statistical error in counting the M peak, which typically contained about 20 000 events, was confined to a few percent and for the L and K peaks, counting was done to within 1% statistical error. The following values were used in Eq. (6): $\omega_K=0.827\pm0.014$ for $Z=47$,[20] $k_\alpha=0.807$,[21] and $k_\beta=0.154$.[21]

Table I summarizes the measurements of the M/L-capture ratio in Cd^{109} decay and includes the averages of 8 runs at 3 atm, 4 runs at 4 atm, and a single run at 5-atm pressure, together with the total K x-ray escape probability P used in Eq. (6). The spread in the P_M/P_L values is relatively small, and the average of the results is

$$P_M/P_L=0.232\pm0.020,$$

where the error is the standard deviation from the mean. This value is compared to the exchange-corrected theoretical value ($P_M/P_L=0.230$) below.

TABLE II. Experimental values of the $(L+M+\cdots)/K$-capture and L/K-capture ratios in Cd^{109} decay and values of Q_{EC} deduced therefrom.[a]

Experimental ratio $P_{L+M+\ldots}/P_K$	P_L/P_K	Method	Ref.	Q_{EC} (keV) to 88-keV level in Ag^{109}
0.24 ±0.04		Scint. spectr.	b	112_{-24}^{+58}
0.42 ±0.18		Scint. spectr.	c	60_{-12}^{+30}
0.332±0.015		Gaseous source in multiwire prop. counter	Present work	78_{-4}^{+11}
	0.30 ±0.04	External source prop. counter	d	66_{-6}^{+10}
	0.28 ±0.03	4π scint. spectr.	e	77_{-4}^{+8}
	0.237±0.015	Gaseous source in multiwire prop. counter	Present work	80_{-3}^{+7}

[a] All values from the literature of Q_{EC} in Table II have been recalculated to include the correction for exchange overlap (see Ref. 4).
[b] A. H. Wapstra and W. van der Eijk, Nucl. Phys. **4**, 325 (1957); **4**, 695 (1957).
[c] A. H. Wapstra, Arkiv Fysik **7**, 265 (1954).
[d] G. Bertolini, A. Bisi, E. Lazzarini, and L. Zappa, Nuovo Cimento **11**, 539 (1954).
[e] E. der Mateosian, Phys. Rev. **92**, 928 (1953).

B. The L/K- and $(L+M+\cdots)/K$-capture Ratios and Transition Energy

The transition energy Q_{EC} for Cd^{109} electron capture to the 88-keV ($\frac{7}{2}+$) state in Ag^{109} has not been measured by either of the two most usual methods, inner bremsstrahlung endpoint or the (p,n) threshold determination. A number of early measurements of the L/K- and $(L+M+\cdots)/K$-capture ratios have been reported, from which Q_{EC} has been deduced. We have corrected these early results for exchange overlap[4] and list the corrected values in Table II, together with the present results. Most of the early values were obtained from K/γ intensity ratios from solid sources using scintillation or external source proportional counting, and thus are subject[2] to fairly large uncertainties arising from the use of K and L fluorescence yields, self-absorption, self-scattering, and self-excitation in the solid sources, and air and window absorption corrections. The present result is not sensitive to these uncertainties, since the source was gaseous and internal.

The $(L+M+\cdots)/K$-capture ratio was measured by operating the counter without anticoincidence, so that the number of K x rays observed is the sum of K x rays resulting from K capture and K x rays arising in the escape of K x rays following K conversion of the 88-keV gamma transition. This depends on the assumption that the K x rays lost from the central counter are replaced by K x rays from the ring counter. (Otherwise, the observed number of K x rays in the central counter would be small, giving a ratio of P_L/P_K of the order of 10 and a N_γ/N_K intensity ratio of about 5, which values contrast sharply with observation.) Further experimental justification for the above assumption was obtained by introducing Fe^{55}-labeled ferrocene vapor into the counter and measuring its L/K-capture ratio at a low pressure of about 0.5 atm. At this low pressure, the fractional escape probability of 6-keV Mn K x rays is approximately 75%, but the L/K-capture ratio[6] remained consistent with the assumption of complete K x-ray replacement. The capture proba-

bility ratio then may be obtained from the equation

$$\frac{P_{L+M+\ldots}}{P_K}=\frac{1+\alpha}{\alpha}\left[\frac{1}{(N_K/N_\gamma)-f_K\omega_K}\right]-1, \quad (9)$$

where α is the total conversion coefficient of the 88-keV gamma transition, and N_γ is the number of internally converted gamma transitions observed in the central counter. The 88-keV unconverted gammas are not detected because of their negligible interaction in the counter gas. The value $\alpha_K=10.3\pm0.5$ was used[19] in the calculation ($\alpha=24$). The substantial uncertainty in α leads to an appreciable uncertainty in the value of $P_{L+M+\ldots}/P_K$, which from the present experiment is 0.332 ± 0.015.

The value of Q_{EC} can be evaluated from the experimental $P_{L+M+\ldots}/P_K$ ratio by means of the equation

$$P_{L+M}/P_K=(\lambda_L/\lambda_K)(q_L/q_K)^2+(\lambda_M/\lambda_K)(q_M/q_K)^2, \quad (10)$$

where (λ_L/λ_K) is the square of the L/K wave function ratio[1] corrected for exchange[4] and has the value 0.1225; (λ_M/λ_K) is the square of the M/K wave function ratio corrected for exchange[7] and has the value 0.024; q_K, q_L, and q_M are the energies of the emitted neutrinos following K, L, and M capture, respectively: and $Q_{EC}=q_i+B_i$, where q_i is the neutrino energy and B_i is the binding energy of the captured electron in the ith shell.

A somewhat more accurate measurement of Q_{EC} is obtained from a measurement of the L/K-capture ratio in which anticoincidence in the ring counter is derived from the converted gamma peak, thereby eliminating that fraction of K x rays from K conversion of the gamma in the ring counter. For this determination one measures N_L/N_K and computes P_L/P_K from

$$P_L/P_K=N_L/N_K-P\omega_K k_\alpha. \quad (11)$$

The value of Q_{EC} is then obtained in a manner similar to that above from the theoretical expression

$$P_L/P_K=(q_L/q_K)^2(\lambda_L/\lambda_K), \quad (12)$$

where $Q_{EC}=q_i+B_i$ as before, and $\lambda_L/\lambda_K=0.1225$. The value of Q_{EC} thus deduced from the experimental value $P_L/P_K=0.237\pm0.015$ (Table II) is

$Q_{EC}=80_{-3}^{+7}$ keV to the 88-keV $(\frac{7}{2}+)$ level in Ag^{109}.

This determination of Q_{EC} is required in order to evaluate the theoretical M/L-capture ratio. From Fig. 1, the ratio of M/L wave functions is 0.195 and the exchange-overlap correction extrapolated from Eq. (2) is $X^{M/L}=1.101$. The factor $(q_M/q_L)^2$ was evaluated using $Q_{EC}=80$ keV, so that the theoretical value for the M/L-capture ratio becomes $(P_M/P_L)=0.230$, within a few percent, which is in excellent agreement with the experimental value, 0.232 ± 0.020 (Table I).

Recently another M/L-capture ratio in this region has been measured. Manduchi and co-workers[22] measured the M/L- and L/K-capture ratios in Sn^{113} decay, with gaseous $Sn^{113}(CH_3)_4$ in a multiwire proportional counter, and found a value of $P_M/P_L=0.20\pm0.01$. The theoretical prediction, 0.22, is some 10% larger. The reason for this discrepancy is at present not understood.

Note added in proof. Additional measurements of the capture ratios in Cd^{109} and Sn^{113} decays have just been reported. Leutz, Schneckenberger, and Wenninger[23] report the following electron-capture ratios in Cd^{109} decay: $(P_L+P_{M,N,\ldots})/P_K=0.228\pm0.003$, $P_L/P_K=0.195\pm0.005$, and $(P_{M+N+\ldots})/P_L=0.17\pm0.05$, from which the transition energy, after correction for exchange overlap, is $Q_{EC}=104\pm4$ keV to the 88-keV level in Ag^{109}. The Cd^{109} source was built into the lattice of NaI(Tl) crystals grown from the melt. In addition, the conversion coefficients for the 88-keV gamma transition were measured: $\alpha_{total}=24.7\pm0.5$, $\alpha_K=11.0\pm0.3$, $\alpha_L=11.7\pm0.8$, and $\alpha_{M,N}=2.0\pm1.1$. The capture ratios are not in good agreement with the present investigation or with the theory. Similarly, from measurements on solid sources of Cd^{109} and Sn^{113} with a special coincidence technique with a semiconductor electron detector and NaI(Tl) scintillator, Etti, Brundrit, and Sen[24] measured the $(P_{L+M+\ldots})/P_K$ capture ratios in Cd^{109} and Sn^{113} decays to be 0.90 ± 0.20 and 0.16 ± 0.02, respectively. These results disagree with those of Leutz *et al.* and with those reported in the present investigation, and, although not corrected for exchange overlap, they disagree strongly with the theory. It is not clear why measurements with nongaseous sources generally do not agree with each other or with the theory. It can be said, however, that the multiwire proportional counter with anticoincidence using gaseous sources generally gives orbital electron-capture ratios which are in good agreement with the theory and which agree well among various independent investigations. For determination of orbital electron-capture ratios, the multiwire counter with gaseous sources remains the only precision method at the present time.

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[22] C. Manduchi, G. C. Nardelli, M. T. Russo-Manduchi, and G. Zannoni, Nuovo Cimento **33**, 49 (1964).
[23] H. Leutz, K. Schneckenberger, and H. Wenninger, Nucl. Phys. **63**, 263 (1965).
[24] D. Etti, D. R. Brundrit, and S. K. Sen (unpublished, and private communication.)