Level Crossings in the $(5s5p)\ ^3P_1$ State of Radioactive Cd^{107} and Cd^{109}†

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Four level crossings, which obey the selection rule $\Delta m_F=2$, have been detected for hyperfine levels in the $(5s5p)^3P_1$ state of both 6.7-h Cd^{107} and 470-day Cd^{109} by observing the change in intensity at 90° of the resonance fluorescence of the 3261 Å intercombination line. The hyperfine interaction constants found from an analysis of the crossing point fields, and corrected for interaction with the adjacent fine structure states of the $5s5p$ multiplet, are $A_{107}=-853.543(6)$ Mc/sec, $B_{107}=-163.279(5)$ Mc/sec, $A_{109}=-1148.784(7)$ Mc/sec, $B_{109}=-165.143(5)$ Mc/sec. The ratios of the magnetic-dipole moments, and the electric-quadrupole moments, to within the effect of hyperfine anomalies, are $\mu_{107}/\mu_{109}=0.742997(8)$; $Q_{107}/Q_{109}=0.98871(4)$.

INTRODUCTION

THE level crossing technique of Colegrove, Franken, Lewis, and Sands[1–3] is a natural complement to double resonance[4] in the investigation of the hyperfine structure (hfs) of the excited states of stable and radioactive atoms. Both techniques are of interest in the study of the nuclear moments and hfs of atoms with diamagnetic $^1S_0$ ground states.

In particular, the lowest lying $(nsnp)\ ^3P_1$ states of the group II metals have lifetimes long enough to permit a precise determination of hfs, while the g_J factors of these states have been measured, or may be calculated accurately enough on the basis of Russell-Saunders coupling, so that the radio frequency $\Delta F=0$, transitions between the Zeeman components of a given hyperfine state, can be found with little searching.[5] While in the limit of low magnetic fields these transitions are insensitive to the hfs of the state, at fields of a few hundred G the interaction with the neighboring hyperfine states is usually sufficient so that a determination of the interaction constants can be made to a percent or better.

To determine the hfs at low fields to the considerably higher precision warranted by the long lifetimes and consequent narrow natural widths of the metastable $^3P_1$ states, the $\Delta F=\pm1$ transitions must be observed in the microwave region. It is simpler in practice, however, to observe the crossing of certain Zeeman components of the hyperfine levels at higher fields. The sensitivity of the level-crossing effect is comparable to double resonance, and the method has the great virtue, in practice, of doing away with all microwave apparatus and requiring only the radio-frequency nuclear magnetic resonance (NMR) measurement of a magnetic field.

If the g_J factor of the state in question is known to high accuracy, for example, from a double-resonance experiment on the even isotopes of the atom, analysis of the observable crossings allows a determination of the magnetic dipole interaction constant A, and the quadrupole interaction constant B, to the same precision that they can be obtained from a $\Delta F=\pm1$ double-resonance experiment.

Against the simplicity of the level-crossing method applied to the Group II metals must be opposed the computational difficulty of evaluating the hyperfine constants from the observed crossings. At low fields the Zeeman interaction between hyperfine levels can be treated by perturbation techniques. Level crossings occur, however, when the Zeeman and hyperfine energies are comparable. This means the diagonal and off-diagonal matrix elements of the Hamiltonian, written in either the $(IJFm_F)$ or (IJm_Im_J) representation, are of the same order of magnitude, and the hyperfine levels can be calculated only by solving the general secular equation. This problem is fortunately amenable to high-speed computing techniques. General programs can be written applicable to a wide variety of hyperfine problems, and in practice the advantages of the level crossing method are found to considerably outweigh its disadvantages.

In this paper we describe the extension of a level crossing experiment already reported[6] for stable Cd^{111} and Cd^{113} ($I=\frac{1}{2}$) to 6.7-h Cd^{107} and 470-day Cd^{109} ($I=\frac{5}{2}$). These radioactive atoms were produced in either case

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by a (p,n) reaction on silver. A method for calculating the hyperfine interaction constants from the observed crossings, taking into account second-order interaction with the adjacent fine structure states, is discussed, and the ratios of the nuclear magnetic-dipole and electric-quadrupole moments of Cd^{107} and Cd^{109} calculated.

ZEEMAN EFFECT AND LEVEL CROSSINGS

Level crossing can be detected by observing a change in the angular distribution of resonance fluorescence when two crossing levels satisfy the selection rule $\Delta m_F=1$, 2.[2] Such crossings have been observed in the Zeeman effect of both the fine structure and the hyperfine structure of atoms.[1,6-9] In general, the number of crossings in the hyperfine structure of a given atom satisfying this rule increases with increasing value of the nuclear spin, and is always sufficient to determine all the hyperfine interaction constants.

For the case $J=1$, $I=\frac{1}{2}$ already studied[6] in $Cd^{111,113}$ there exists only one $\Delta m_F=2$, and no $\Delta m_F=1$ crossing. For $I>\frac{1}{2}$ there are in general $(2I-1)$ of the $\Delta m_F=2$ and $(2I-1)$ of the $\Delta m_F=1$ crossings. For spectroscopic purposes the $\Delta m_F=2$ crossings are of the greater interest

TABLE I. Hyperfine-interaction constants for the $(5s5p)^3P_1$ state, and spins and nuclear magnetic-dipole moments of Cd^{107} and Cd^{109}, as determined by low-field $\Delta F=0$ double resonance (Refs. 5 and 10).

	Cd^{107}	Cd^{109}
A (Mc/sec)	$-854.2(10)$	$-1148.6(20)$
B (Mc/sec)	$-166.0(30)$	$-167.3(20)$
I	$\frac{5}{2}$	$\frac{5}{2}$
μ	$-0.6162(8)\mu_N$	$-0.8286(15)\mu_N$

since they can be observed without polarization of the incoming or scattered light, and have a Lorentzian line shape when the scattered light, the incident light, and the applied field are mutually perpendicular.[2,6]

Prior to the present work, $\Delta F=0$ double-resonance experiments had established the spins of $Cd^{107,109}$,[5,10] and the hyperfine constants of the $(5s5p)^3P_1$ state, with sufficient accuracy to allow location of the crossing fields to about a tenth of 1%. A summary of the double-resonance results is given in Table I.

The Zeeman effect for Cd^{107}, calculated on the basis of the Hamiltonian

$$\mathcal{H}=A\mathbf{I}\cdot\mathbf{J}+\frac{B}{2I(2I-1)J(2J-1)}\times[3(\mathbf{I}\cdot\mathbf{J})^2+\tfrac{3}{2}(\mathbf{I}\cdot\mathbf{J})-I(I+1)J(J+1)]+g_J\mu_0\mathbf{J}\cdot\mathbf{H}+g_I\mu_N\mathbf{I}\cdot\mathbf{H}, \quad (1)$$

is shown in Figs. 1 and 2.

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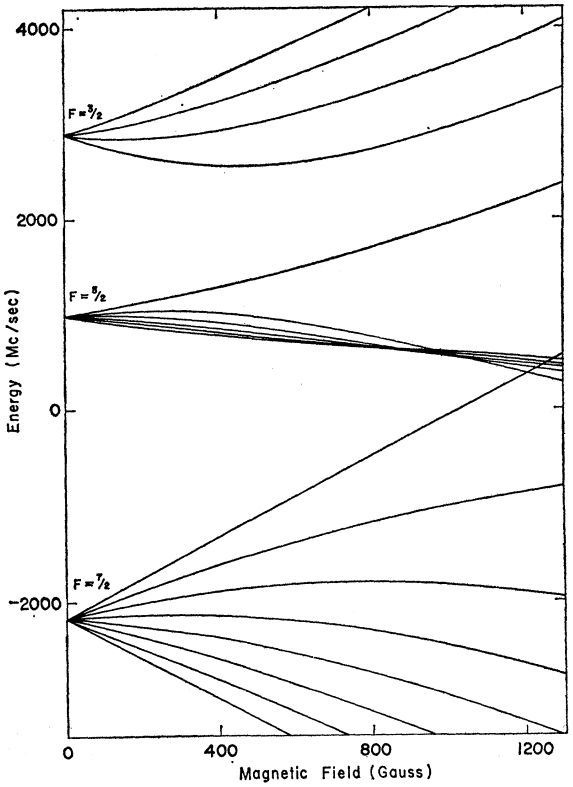

FIG. 1. Zeeman effect of the $(5s5p)^3P_1$ state of Cd^{107}.

It is seen that of the four $\Delta m_F=2$ crossings there is only one for which $\Delta F=1$. This is true for all I, this crossing being that of the $F=I+1$, $m_F=\pm(I+1)$, and the $F=I$, $m_F=\pm(I-1)$ levels. The sign to be chosen is the opposite of that of the magnetic interaction constant A.

The $\Delta F=1$ crossing is the most intense and the

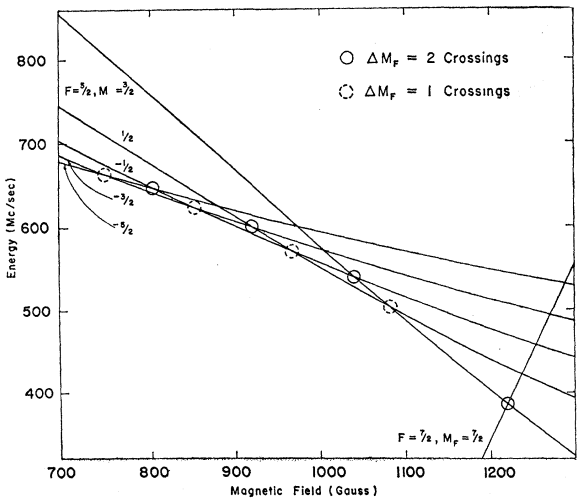

FIG. 2. Details of the level crossings in the $(5s5p)^3P_1$ state of Cd^{107}.

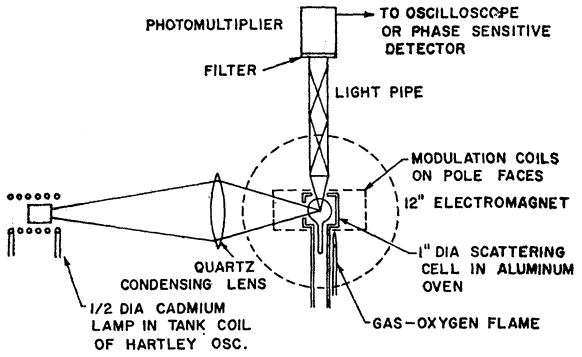

FIG. 3. Schematic illustration of the experimental apparatus.

sharpest observed. Its position is insensitive to the quadrupole interaction constant B, and it may therefore be used to establish the ratio A/g_J to high precision.

The $(2I-2)$ in number, $\Delta F=0$ "foldover" crossings, however, for which $F=I$, lie at fields which are highly sensitive to B. Due to the small relative slope, $dW_1/dH - dW_2/dH$, these crossings are observed to be much broader than the $\Delta F=1$ main crossing. They are also observed to be much less intense, so that the crossing fields can be measured with considerably less precision than for the main crossing. Experience has shown, however, that they are so sensitive to B that collectively they allow B to be found to the same accuracy as A.

LEVEL CROSSING EXPERIMENT

A. Preparation of Radioactive Sample

Double-resonance and level-crossing experiments are capable of great sensitivity, due to the relatively small effect of thermal noise, and the large effective resonance-scattering cross sections of atoms in the optical region. Since effectively all the isotopes of a given atom contribute to resonance fluorescence, it is important that the particular isotope investigated have as large a fractional abundance as possible.[5] When radioactive atoms are studied, it is of particular concern that stable impurities be reduced as far as possible.

Cd^{107} and Cd^{109} were produced by a (p,n) reaction on natural silver (specified to contain less than one part in 10^7 cadmium impurities) at an energy for the bombarding protons near 11 MeV. Ag^{107} and Ag^{109} have a natural abundance of 51.35 and 48.65%, respectively, and nearly equal cross sections for the (p,n) reactions over the energy range in question,[10] so that nearly equal numbers of the two species of radioactive atoms resulted. A bombardment of about 400 μA-h was found to yield about 0.4 C of Cd^{107}, or about 5×10^{14} atoms. If care was taken in the preparation of the scattering cell, it was found that the $Cd^{107,109}$ crossings were considerably stronger than those of stable $Cd^{111,113}$, indicating that the total number of stable atoms was less than the number of radioactive atoms (see below).

The scattering cell was identical to that used in the low-field double-resonance experiments. Further details of the production of the radioactive atoms, and the preparation and filling of the cells may be found in Refs. 5 and 10.

B. Measurements

The level crossings in Cd^{107} and Cd^{109} were observed, and the crossing-point magnetic fields measured, with the same apparatus used for Cd^{111} and Cd^{113}, shown in Fig. 3. The rf electrodeless resonance lamp consisted of a small quartz cylinder, 1.5 cm in diameter and 2.5 cm long, which was thoroughly baked out and purged with an rf electric discharge, evacuated to better than 10^{-7} Torr, and filled with distilled cadmium and one or two Torr of spectroscopically pure argon. The four $\Delta m_F=2$ crossings for Cd^{107} are shown in Fig. 4. Even if the scattering cell was prepared in a time short compared to the half-life of Cd^{107} (6.7 h), the Cd^{109} crossings were observed to be somewhat stronger than those of Cd^{107}, presumably due to greater illumination of the Cd^{109} crossings by the natural cadmium lamp. A similar effect was observed previously for Cd^{113} versus Cd^{111}.[6] As expected, the Cd^{107} crossings faded over a period of several half-lives.

The $\Delta F=1$ crossing for either isotope was strong

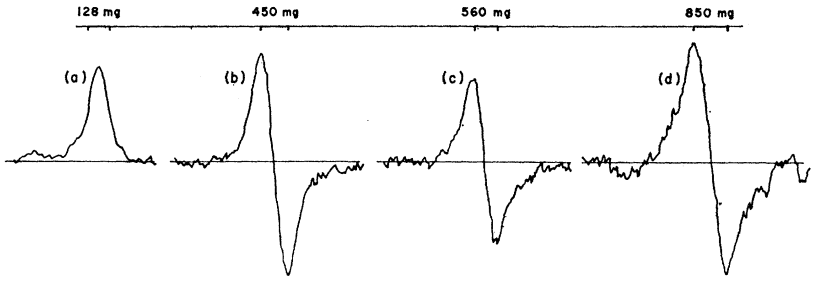

FIG. 4. Observed level crossings for Cd^{107}. (a) is an oscilloscope trace of the main $(\frac{7}{2},\frac{7}{2})$, $(\frac{5}{2},\frac{5}{2})$ crossing. (b), (c), and (d) are recorder traces with phase-sensitive detection of the $\Delta F=0$ crossings: $(\frac{5}{2},\frac{5}{2})$, $(\frac{5}{2},-\frac{1}{2})$; $(\frac{5}{2},\frac{1}{2})$, $(\frac{5}{2},-\frac{3}{2})$; and $(\frac{5}{2},-\frac{1}{2})$, $(\frac{5}{2},-\frac{5}{2})$, with time constants of 1, 3, and 10 sec, respectively. The vertical scale is arbitrary in each case. (a) has a Lorentzian line shape, while (b), (c), and (d) have line shapes, due to phase-sensitive detection, which are the derivative of a Lorentzian.

enough to be observed with oscilloscope display, and the crossing field was measured as for $Cd^{111,113}$, by superimposing the NMR signal of protons in mineral oil and the level crossing signal using a dual beam oscilloscope. The three $\Delta m_F=2$, $\Delta F=0$ crossings for either isotope were too weak to be studied in this way, but were observed with good signal strength with phase-sensitive detection and time constants from 1 to 10 sec. A comparison of the observed signal strengths, and those calculated on the basis of Eq. (2) of Ref. 6, and the assumption of equal illumination of all four crossing points, is given in Table II. The measured crossing point magnetic fields are given in Table III.

The crossing signals of stable Cd^{111} and Cd^{113}, each about 12% abundant, could always be observed in the same cell with the crossings of the radioactive atoms, due probably to cadmium impurities in the silver target. For targets which were weakly bombarded, the signal strength for these crossings was about the same as that observed previously,[6] indicating that the majority of the resonance fluorescence was due to the stable isotopes. For strongly bombarded targets, however, the $Cd^{111,113}$ signals were weak, and the $\Delta F=1$ crossing for Cd^{107} and Cd^{109} was several times stronger than the strongest signals ever observed for $Cd^{111,113}$, indicating that practically all of the fluorescence was due to the radioactive atoms.

TABLE II. Calculated and observed relative intensities of the crossings in Cd^{107} and Cd^{109}, normalized to 1.00 for the $(\frac{5}{2},\frac{3}{2})$, $(\frac{5}{2},-\frac{1}{2})$ crossing.

Crossing levels, (F_1,m_1) (F_2,m_2)	Cd^{107} calc.	Cd^{107} obs.	Cd^{109} calc.	Cd^{109} obs.
$(\frac{7}{2},\frac{7}{2})$, $(\frac{5}{2},\frac{3}{2})$	7.55	11.0	7.74	11.6
$(\frac{5}{2},\frac{3}{2})$, $(\frac{5}{2},-\frac{1}{2})$	1.00	1.0	1.00	1.0
$(\frac{5}{2},\frac{1}{2})$, $(\frac{5}{2},-\frac{3}{2})$	0.69	0.47	0.67	0.48
$(\frac{5}{2},-\frac{1}{2})$, $(\frac{5}{2},-\frac{5}{2})$	0.31	0.22	0.28	0.37

HYPERFINE CONSTANTS

On the basis of Eq. (1) four constants, A, B, g_J, g_I, determine the hyperfine structure and the Zeeman effect of the $(5s5p)\,^3P_1$ state, or conversely might be expected to be found from an analysis of the level crossings. Since $g_I\mu_N$ is small with respect to $g_J\mu_0$ however, and it is known for either isotope from the double-resonance experiments to well within 1%, it is considered a constant as regards the fourth term of Eq. (1) in the present calculation. Moreover, g_J is already known to 6 ppm from the level crossing experiment on $Cd^{111,113}$,[6] and, as will be shown below, is not in any case determined to high precision by the level crossings. A and B are, therefore, overdetermined by the four observed $\Delta m_F=2$ crossings.

A least-square fitting of the calculated to the observed crossings was performed with an IBM-7090 computer.

TABLE III. Measured values of the crossing fields in units of the nuclear magnetic resonance frequency of protons in mineral oil. The uncertainties given are standard deviations plus an estimated contribution of 15 cps for possible systematic errors.[a]

Crossing levels, (F_1,m_1) (F_2,m_2)	Cd^{107} crossing fields (kc/sec)	Cd^{109} crossing fields (kc/sec)
$(\frac{7}{2},\frac{7}{2})$, $(\frac{5}{2},\frac{3}{2})$	5189.528(20)	6987.010(20)
$(\frac{5}{2},\frac{3}{2})$, $(\frac{5}{2},-\frac{1}{2})$	4412.103(49)	6177.579(78)
$(\frac{5}{2},\frac{1}{2})$, $(\frac{5}{2},-\frac{3}{2})$	3898.517(52)	5593.874(55)
$(\frac{5}{2},-\frac{1}{2})$, $(\frac{5}{2},-\frac{5}{2})$	3410.542(85)	5007.918(90)

[a] Preliminary results have appeared in Bull. Am. Phys. Soc. 7, 25 and 433 (1962).

A program was already available[11] to conduct such an analysis on the basis of the Hamiltonian of Eq. (1). Second-order corrections, however, due to interaction with the other fine-structure states of the $(5s5p)$ multiplet, contribute fractional corrections to A and B of the order of the ratio of the hyperfine or Zeeman energies to the fine-structure separations. This is of the order of 100 ppm in the present case, and is much larger than experimental uncertainties. The existing program was therefore modified to add these corrections, the terms off diagonal in J being calculated from the paper of Lurio, Novick, and Mandel,[12] and folded into the $(5s5p)\,^3P_1$ state by the transformation method of Van Vleck.[13]

The interaction constants which gave the best fit to the observed crossings are listed in Table IV. The "zero-field" constants are those which would be determined directly from a low-field double-resonance experiment—only second-order corrections proportional to the first and second power of the applied field were retained in the fitting calculation. In the calculation of the "isolated" constants, however, second-order hyperfine corrections independent of the field were included. These are then the constants which would be found from a low-field double-resonance experiment if second-order hyperfine corrections were made, and are those from which the nuclear moments should be calculated.

The constants in Table IV were calculated taking $g_J=1.499850$, a weighted average from the reported level crossing experiment on $Cd^{111,113}$, and the results of a high-field double-resonance experiment on the even

TABLE IV. Hyperfine-interaction constants of the $(5s5p)^3P_1$ state of Cd^{107} and Cd^{109}.

	A_{107} (Mc/sec)	B_{107} (Mc/sec)	A_{109} (Mc/sec)	B_{109} (Mc/sec)
Low field	−853.583(6)	−163.606(5)	−1148.852(7)	−165.730(5)
Isolated	−853.543(6)	−163.279(5)	−1148.784(7)	−165.143(5)

[11] Kindly furnished by D. H. Zurlinden, Lawrence Radiation Laboratory, University of California, Berkeley, California.

[12] A. Lurio, M. Mandel, and R. Novick, Phys. Rev. 126, 1758 (1962).

[13] E. C. Kemble, *Fundamental Principles of Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1937), p. 394.

cadmium isotopes.[14] It was found, as expected, that an equally good fit of the calculated to the observed crossings could be obtained if g_J was selected to lie within a few percent of this value. Level crossing experiments therefore, in fact, determine the ratio of the hyperfine interaction constants to the g_J factor of the state, and the g_J factor must be established by other means if the hyperfine constants are to be found to high precision.

NUCLEAR MOMENTS

The hyperfine anomaly for $Cd^{111,113}$ ($I=\frac{1}{2}$) has been shown to be only 1.6×10^{-5} from an atomic-beam investigation of the $^3P_2$ state.[15] It is not anticipated, however, that this effect will be so small for isotopes with different spins. For the cases of Hg^{199} ($I=\frac{1}{2}$) and Hg^{201} ($I=\frac{3}{2}$), and Xe^{129} ($I=\frac{1}{2}$) and Xe^{131} ($I=\frac{3}{2}$), also even-proton, odd-neutron nuclei, hyperfine anomalies of, respectively, 0.17 and 0.044% have been measured.[16,17]

We, therefore, cannot expect to determine the individual magnetic dipole moments of $Cd^{107,109}$ substantially more accurately from the known moment[18] of Cd^{111}, and the ratios of the A constants, than from the low-field double-resonance experiments. If we assume that the hyperfine anomaly of $Cd^{107,109}$ is comparable to that of $Cd^{111,113}$, however, we may calculate the ratio of the magnetic dipole moments to be

$$\mu_{107}/\mu_{109}=A_{107}/A_{109} =0.742997(8). \quad (2)$$

To within the effect of hyperfine anomaly we may likewise calculate the ratio of the electric quadrupole moments to be

$$Q_{107}/Q_{109}=B_{107}/B_{109}, =0.98871(4). \quad (3)$$

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Level Structure of Ni^{64}

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Excited states of Ni^{64} have been investigated through the study of the inelastic scattering of 11-MeV protons from an isotopically enriched target of Ni^{64}. Excitation energies of 30 levels between 1.34 and 5.49 MeV are reported. For the level at 3.55 MeV a spin and parity assignment of 3^- is suggested.

THE excited states of Ni^{64} have been studied by measuring the inelastic scattering of protons from a Ni^{64} target, and the results have been compared with those recently reported by Benveniste, Mitchell, and Fulmer.[1] The Ni^{64} target was a self-supporting metal foil, ~1 mg/cm^2, enriched to 99.8% in the isotope. An 11-MeV proton beam from the ORNL Tandem Van de Graaff impinged on the target, and pulse-height spectra of the scattered protons were obtained at angles of 60, 80, 100, and 120 deg from the beam axis. The scattered protons were detected with surface-barrier silicon detectors, the over-all energy resolution being ~40 keV (full width at half-maximum).

Proton groups scattered by nickel were distinguished from those scattered by other nuclei (impurities) in the target and from alpha groups resulting from the (p,α) reaction by the characteristic change in energy with the angle of observation. To further distinguish between alpha and proton groups, a 2-mg/cm^2 aluminum absorber was placed between the target and the counter during one of two measurements at 80 deg.

The pulse-height spectrum obtained at 60 deg is shown in Fig. 1. Protons scattered by the first excited state of Fe^{56} ($Q=-0.845$ MeV) are clearly evident in this figure and indicate an iron contamination of

* Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.
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