Nuclear Spins of Six Neutron-Deficient Gold Isotopes*

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(Received July 5, 1960)

The nuclear spins of six neutron-deficient isotopes of gold have been measured by atomic-beam magnetic-resonance methods. The results of these measurements are as follows: Au^{191} (3.0 hr), $I=\frac{3}{2}$; Au^{192} (4.8 hr), $I=1$; Au^{193} (17.5 hr), $I=\frac{3}{2}$; Au^{194} (39 hr), $I=1$; Au^{195} (190 day), $I=\frac{3}{2}$; Au^{196} (5.6 day), $I=2$.

I. INTRODUCTION

An atomic-beam magnetic-resonance apparatus of the flop-in type has been used to measure the nuclear spins of six neutron-deficient isotopes of gold. The apparatus has been described before.[1] Discussion of the experimental method will be limited to those changes in apparatus or procedure that have been made necessary by the use of nonalkali beams.

The isotopes used in the experiments were produced in the Crocker 60-in. cyclotron on the Berkeley campus of the University of California. This cyclotron is capable of producing beams of protons, deuterons, or alpha particles with energies of ~12 Mev/nucleon. The alpha and deuteron beams have been used exclusively in these experiments. Alpha bombardment of iridium targets was used to produce those isotopes between Au^{191} and Au^{194}. Although the same type of bombardment could be used to make Au^{195} and Au^{196}, these two isotopes were usually produced from deuteron irradiation of platinum. Because of the longer half-lives of the heavier gold isotopes, it is necessary to have more atoms of each isotope in order to obtain a given amount of radioactivity. Platinum foil could be used as an internal target in the cyclotron, where the higher beam density made it possible to make larger amounts of the gold isotopes. Furthermore, the methods of separation of gold from the target material were much more efficient when the target material was platinum.

Since the principal decay mode for each isotope is by electron capture, small crystal scintillation counters could be used to detect efficiently the x rays of any of the isotopes. Except for Au^{191} and Au^{192} (which cannot have the same spin) the half-lives of the isotopes differ significantly from each other. A crude decay curve of each spin sample thus positively identifies the isotope involved.

The results for Au^{191}, Au^{193}, and Au^{195} confirm the ground-state assignment of the seventy-ninth proton to the $2d_{\frac{3}{2}}$ level.[2] It is more difficult to assign single-particle levels to the odd neutrons in those gold isotopes with even mass numbers.

II. EXPERIMENT

The standard atomic-beam "spin-search" experiment for an atom with $J=\frac{1}{2}$ consists of a search through a (usually small) number of radio frequencies for the one which is able to cause a transition between the energy levels corresponding to $F=I+\frac{1}{2}$, $M_F=-I-\frac{1}{2}$, and $F=I+\frac{1}{2}$, $M_F=-I+\frac{1}{2}$. At low magnetic fields, the frequency of this transition is related to the nuclear spin by the simple equation

$$\nu=-g_J\frac{\mu_0 H}{h}\frac{1}{2I+1}.$$

In a flop-in apparatus,[3] the occurrence of such a transition is accompanied by an increase in the number of atoms that are able to reach the detector. In order to be able to choose the proper frequencies for a spin search, it is necessary to know the Lande g factor for the atom and the strength of the applied magnetic field. For the gold isotopes, the atomic g factor for the $^2S_{\frac{1}{2}}$ ground state, as measured by Wessel and Lew, is $g_J=-2.00412\pm0.00012$.[4] The magnetic field is calibrated by means of a beam of alkali atoms, the nuclear and electronic constants of which are known. This beam originates from a source which is placed behind the source of radioactive gold atoms. When a calibration is desired, the oven containing the gold is moved aside, allowing the alkali beam to pass through the apparatus. A surface ionization detector and associated electrometer make possible a field calibration within a few minutes.

The source of radioactive gold atoms used here was machined from a $\frac{3}{8}$-in. cube of tantalum metal. On the front of this oven, a pair of jaws could be adjusted to produce a beam a few thousandths of an inch wide. The oven was loaded from the top, after which a press-fitting cap was inserted. The oven was heated in the apparatus by electron bombardment to produce a satisfactory beam of gold atoms.

Since the beam was to be detected by the presence of radioactive atoms, suitable collectors were devised. These are in the form of sulfur-coated brass "buttons." The sulfur provides a surface upon which the gold atoms

* This research was supported jointly by the U. S. Atomic Energy Commission and the Office of Naval Research.

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[1] J. P. Hobson, J. C. Hubbs, W. A. Nierenberg, H. B. Silsbee, and R. J. Sunderland, Phys. Rev. **104**, 101 (1956).

[2] M. G. Mayer and J. H. D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley & Sons, New York, 1955).

[3] J. R. Zacharias, Phys. Rev. **61**, 270 (1942).

[4] G. Wessel and H. Lew, Phys. Rev. **92**, 641 (1953).

may stick while they are being counted. The brass button which contains the sulfur gives mechanical strength to the detector samples, as well as providing a convenient and reproducible way of exposing the sulfur surface to the beam and of presenting the collected gold atoms to the detector crystal for counting.

After a timed exposure to the radioactive beam under controlled conditions, each sample is removed from the beam apparatus through a vacuum interlock. The sulfur surface is protected by a piece of cellophane tape, and the button is removed to the counting room. As frequently as is practical, the counting rate on the sample is measured, and a decay curve is generated. The composition of each sample can be determined from a knowledge of the half-lives of the various isotopes that may be present in the sample. An analysis of each decay curve can be made graphically or numerically. A numerical process has been devised for use with the IBM 650. From a knowledge of the counting rates of a sample at various times, along with a list of the different isotopes that may participate in the decay, the computer calculates the composition of each sample at some common time. Comparison of this initial composition for several spin-resonance exposures provides knowledge of which isotope, if any, is participating in a given resonance. This knowledge leads directly to a spin assignment for each radioactive isotope.[5]

If there are appreciable fluctuations in the intensity of the radioactive beam, it becomes necessary to apply some correction factor to each exposure to "normalize" the counting rates to what they would have been if all exposures had been made to the same integrated beam intensity. An ideal normalization procedure would involve the measurement of some fixed fraction of the radioactivity that left the oven during the resonance exposure. No provision has been made in the present apparatus for such a measurement. However, by similiar measurements before and after each resonance exposure, satisfactory normalization factors can be interpolated. The fraction of the beam that is monitored consists of those atoms whose velocities are high enough that they are able to pass through the magnetic fields of the apparatus without suffering a certain minimum deflection. For a beam of gold atoms, this fast fraction may contain as much as 50% of the atoms which make up the beam. During a resonance exposure, the fast fraction is intercepted by a "stop wire," whose geometric shadow shields the detector from the source of radioactive atoms. When the stop wire is removed and the rf field turned off, only the fast fraction of the beam is detected during a "half-beam" exposure (a traditional misnomer which is nearly correct for the gold isotopes).

The usual method of normalization is to plot a curve of half-beam intensity (corrected for radioactive decay) as a function of time. From such a curve, the "half-beam" intensity may be obtained for each resonance exposure for subsequent normalization.

III. ISOTOPE PRODUCTION AND EXPERIMENT

A. Platinum Target

The deuteron bombardment of platinum was used to produce Au^{194}, Au^{195}, and Au^{196}. The expected trace amount of Au^{192} was not observed, although no attempt was made to perform the experiment with sufficient speed to detect this isotope. Nor was the 10-hr isomer of Au^{196} observed during these experiments.[6] Since the x-ray counters are quite selective, no appreciable interference was observed from the β-unstable Au^{198} or Au^{199}.

For irradiation in the cyclotron, the platinum target was usually a piece of foil $1\frac{1}{2}$ by $1\frac{3}{4}$ by 0.020 in. Since the cyclotron beam struck only about half of this target, some trimming could be done before the chemical separation was performed. The platinum target was dissolved along with a weighed amount of stable gold carrier in hot, concentrated aqua regia. The resultant solution was evaporated just to dryness, and the residue taken up in dilute HCl. Addition of ethyl acetate resulted in the extraction of gold choride into a layer above the platinum-bearing HCl. After separation, purification, and drying of the acetate fraction, the residue was taken up in dilute HCl. A jet of SO_2 gas was directed across the surface of this solution, causing the reduction of gold to the metal. This gold was then dried and loaded into the atomic-beam oven.

B. Iridium Target

The iridium target used here was in the form of small pieces of 0.006-in. sintered foil contained behind three 0.001-in. aluminum foils. This thickness of iridium is sufficient to stop the 48-Mev alpha beam, so that all isotopes wlth $191 \leq A \leq 196$ were produced. In practice, the bombardments were short enough that the longer lived isotopes Au^{195} and Au^{196} were not produced in significant quantities. Again, the 10-hr isomer of Au^{196} was not observed. The (α,n) cross section is quite small, of course, compared to those reactions that produce the other gold isotopes.

After irradiation, the iridium pieces were loaded into an atomic-beam oven and heated. When the temperature became high enough, the gold was observed to diffuse out of the target material at a rate sufficient to be used in experiments. Even at the highest temperatures used here, the iridium foil was not melted. Consequently, it could be recovered as foil and reused in later

[5] It should be remarked that spin assignments are actually made to isotopes that have certain half-lives rather than to isotopes of certain mass numbers. The correlations between half-lives and isotopes have been made previously, and the certainty of each spin assignment rests on previous work in the study of radioisotopes. Unless otherwise noted, half-life information is taken from the tables of D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).

[6] R. van Lieshout, R. K. Girgis, R. A. Ricci, A. H. Wapstra, and C. Ythier, Physica **25**, 703 (1959).

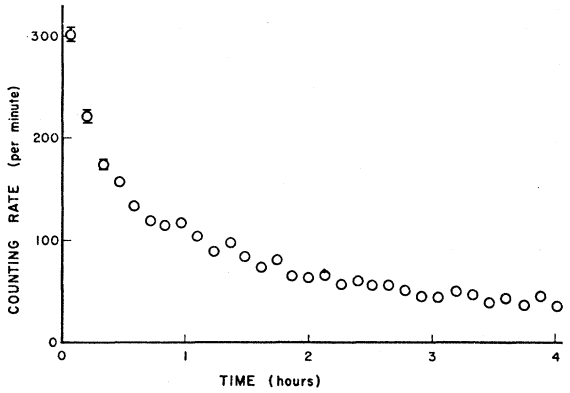

FIG. 1. Decrease in half-beam intensity as radioactive gold is diffused out of an iridium target. Each point represents a 1-min exposure to the fast fraction of the beam.

bombardments. Since iridium is virtually insoluble in the standard reagents, it is fortunate that satisfactory beams of gold can be obtained in the above way without complicated procedures. Although the recovery efficiency of the method has been estimated at only about 50%, the increased speed with which a beam can be produced provides considerable compensation.

The radioactive beams produced by the thermal-diffusion method are never so stable as those obtained from an oven containing a pure metal. However, the instability is almost predictable, so that a simple normalization procedure is possible. Figure 1 shows the variation with time of the counting rate observed on a set of half-beam exposures. A smooth curve can be drawn through these points, and the normalization factor for each resonance exposure is easily determined.

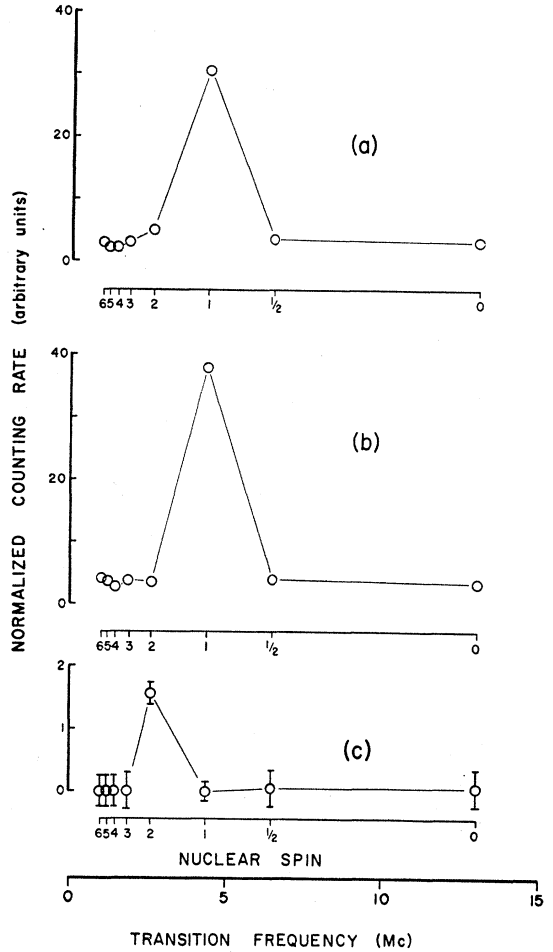

FIG. 2. Spin search at $H=4.65$ gauss for Au^{194} and Au^{196} produced by Pt(d,kn)Au reactions. (a) normalized initial counting rates; (b) normalized 39-hr fraction for each point of (a), establishing $I=1$ for Au^{194}; and (c) normalized 5.6-day fraction of (a), establishing $I=2$ for Au^{196}.

IV. RESULTS: NUCLEAR SPINS

A. Au^{194} and Au^{196}

The normalized results of a search of integral spins using gold separated from platinum are shown in Fig. 2(a). The platinum probe had been bombarded with 367 μa-hr of 24-Mev deuterons over a period of 4.6 hr. With the transition field set at 4.65 gauss, timed exposures were made at frequencies corresponding to all integral spin values between $I=0$ and $I=6$. Each of the samples was counted periodically during the succeeding 5 days. The two samples which had appreciable activity after this time, ($I=1$ and $I=2$) were counted for another 10 days. Each decay curve was then analyzed into its 39-hr and 5.6-day components. The results of this analysis are displayed in Figs. 2(b) and 2(c). From this evidence, spin values of $I=1$ for 39-hr Au^{194} and $I=2$ for 5.6-day Au^{196} were assigned. The spin of Au^{194} has been previously reported.[7]

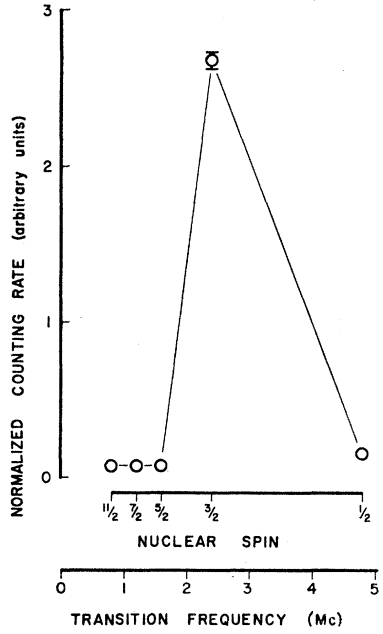

FIG. 3. Spin search at $H=3.43$ gauss establishing $I=\frac{3}{2}$ for Au^{195}.

[7] J. B. Reynolds, R. L. Christensen, W. M. Hooke, D. R. Hamilton, H. H. Stroke, W. B. Ewbank, W. A. Nierenberg, H. A. Shugart, and H. B. Silsbee, Bull. Am. Phys. Soc. 2, 317 (1957).

Confirmation resonances have been observed and the hyperfine structure studied at frequencies up to 40 Mc/sec for Au^{196} and 820 Mc/sec for Au^{194}. These experiments will be summarized in a later article.

B. Au^{195}

Because of the very long half-life,[8] it is difficult to produce sufficient quantities of Au^{195} to give enough activity for a measurement. However, by using long deuteron bombardments accumulated over a period of several weeks, the experiment was made possible. After the last irradiation, the target was stored to allow all shorter activities to decay away. Approximately a year later, the target was dissolved and a spin search done with the activity that remained. The results of this spin-search are shown in Fig. 3. Of the five possibilities, only $I=\frac{3}{2}$ shows a resonance. Since Au^{195} should be the dominant component of the gold sample, a spin of $I=\frac{3}{2}$ was assigned to Au^{195}. Further spin resonances have been obtained at frequencies up to 210 Mc/sec.

C. Au^{191}, Au^{192}, Au^{193}, and Au^{194}

Alpha irradiation of iridium foil has been used to produce the shorter lived gold activities: Au^{191} (3.0-hr), Au^{192} (4.8-hr), Au^{193} (17.5-hr), and Au^{194} (39-hr). The fact that Au^{194} was also produced from this bombardment provided an independent check on its spin measurement ($I=1$) described above.

Normalized results of a spin search using radioactive gold produced from iridium are shown in Fig. 4. This

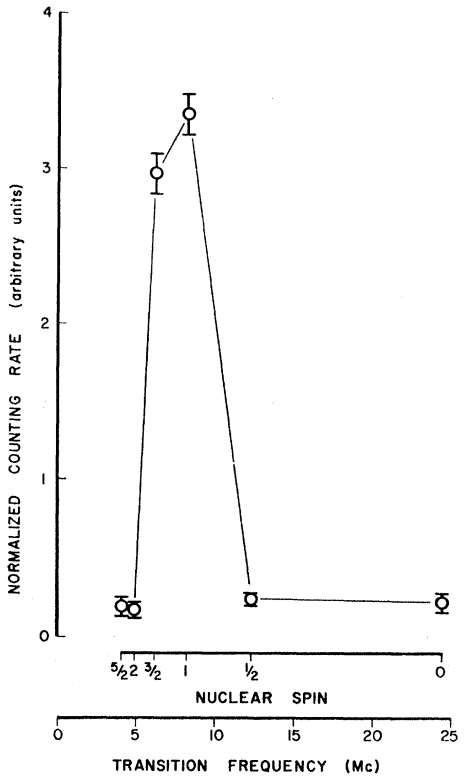

FIG. 4. Spin search at $H=8.7$ gauss for Au^{191}, Au^{192}, Au^{193}, and Au^{194} produced by Ir(α,kn)Au reactions. The data shown are from the first count only.

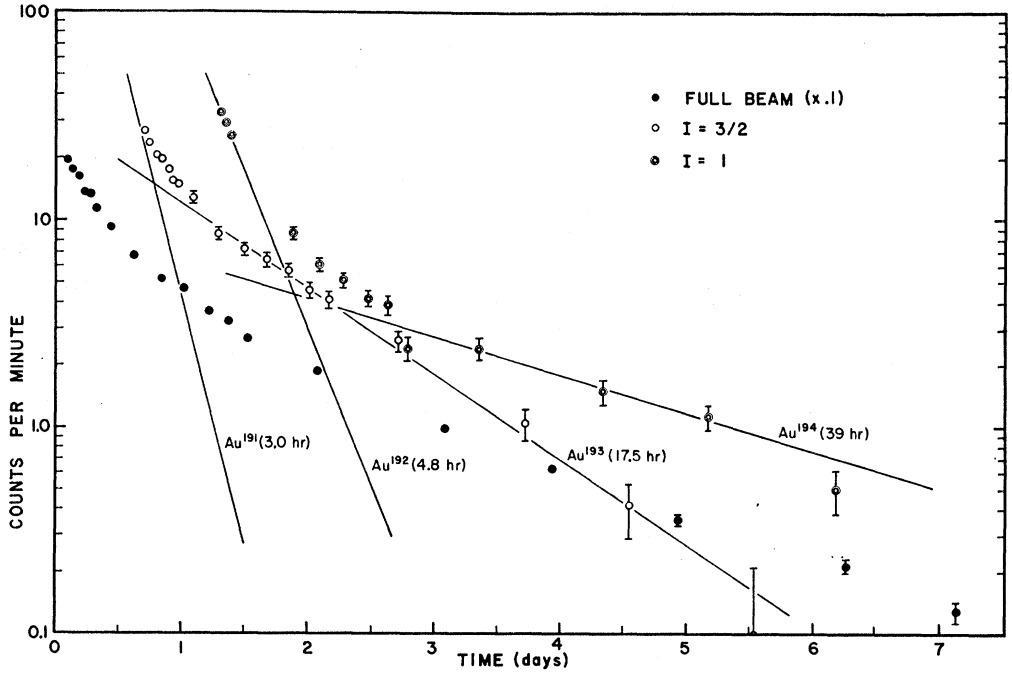

FIG. 5. Identification by decay analysis of gold spin resonances shown in Fig. 4. The time scale has been shifted for the spin-resonance samples in order to provide a clear separation of the decay curves.

[8] A. Bisi, E. Germagnoli, and L. Zappa, Nuovo cimento **11**, 843 (1959).

TABLE I. Composition by activity of Au spin-resonance exposures.

Sample	Isotope Au^{191}	Au^{192}	Au^{193}	Au^{194}
$I=1$	0 (11)	32 (10)	3 (2)	4.3 (6)
$I=\frac{3}{2}$	16 (6)	1 (6)	16 (2)	0.6 (5)
Full-beam	48 (14)	119 (13)	60 (3)	25.2 (9)

spin search was made at a field of 8.7 gauss. The only apparent resonances occurred at frequencies corresponding to $I=1$ and $I=\frac{3}{2}$. Assignment of isotopes to these resonances was made after an analysis of the decay curves of the two high-spin buttons and a sample of the radioactive beam from the same oven. These decay curves are drawn in Fig. 5. Each of the spin-resonance exposures corresponding to $I=1$ and $I=\frac{3}{2}$ has a short-lived component with a half-life of a few hours. The two decay curves then change slope quite differently, with $I=1$ going into the 39-hr decay and $I=\frac{3}{2}$ going into the 17-hr decay.

Digital analysis of these decay curves gave the normalized compositions which are compared in Table I. The number in parentheses represents the uncertainty in the least significant digit of each counting rate. The comparatively large uncertainties on the amounts of Au^{191} and Au^{192} in each sample are due to the similarity in half-lives of the two isotopes. It is apparent that four resonances have been observed here, leading to spin assignments of $I=1$ to Au^{192} and Au^{194}, and $I=\frac{3}{2}$ to Au^{191} and Au^{193}. These assignments have been confirmed by resonances observed up to fields of several hundred gauss. Resulting hyperfine-structure measurements will be reported later. The spin measurements on these four isotopes have been reported previously.[7,9]

V. REMARKS

The assignment of $I=\frac{3}{2}$ to Au^{191}, Au^{193}, and Au^{195} presents no new problems in level-structure interpretation of the gold isotopes. Previous measurements on Au^{197} and Au^{199} have led to the assignment of the seventy-ninth proton to the $2d_{\frac{3}{2}}$ level on the basis of the simple shell model.[2,10,11] The present measurements add further weight to this assignment.

TABLE II. Possible spin values for the odd-odd isotopes of gold, using single-particle levels.

Neutron level	Proton level $2d_{\frac{3}{2}}$	$3s_{\frac{1}{2}}$	$2d_{\frac{5}{2}}$	$1h_{11/2}$
$3p_{\frac{3}{2}}$	2, (1)	**0**	**2**	**5**
$3p_{\frac{1}{2}}$	**0**	2, (1)	4, ···2, (1)	7, ···5, (4)
$2f_{\frac{5}{2}}$	4, ···2, (1)	**2**	**0**	**3**
$1i_{13/2}$	5	7, (6)	9, ···5, (4)	12, ···2, (1)

A nuclear spin of $I=2$ obtained here for Au^{196} and previously[11] for Au^{198} is easily explained from the coupling rules of Nordheim.[12] Several possible couplings can be used to obtain a spin of 2, perhaps the most plausible of which is that suggested by Christensen *et al.*[11] The $3p_{\frac{3}{2}}$ neutron observed in Pt^{195}, Hg^{197}, and Hg^{199} was coupled to the $2d_{\frac{3}{2}}$ proton of gold to give a spin of 2 and reasonable agreement with the measured nuclear moment of Au^{198}.

If Nordheim's rules are strictly applied, there is no simple nuclear configuration that will couple to a spin of 1. The possible level assignments for the odd neutron and odd proton are given in Table II, along with the various spin values to which the particles may be expected to couple. Those spin values in bold-face type indicate applications of Nordheim's strong rule, while other combinations are obtained from the weak rule. Parentheses have been used to mark those spin values obtained by assuming a coupling to $I=|j_n-j_p|$ in cases where the weak rule ($I>|j_n-j_p|$) should be applicable. This assumption corresponds to the modification of Nordheim's rules which has been suggested recently by Brennan.[13] The fact that a spin of 1 cannot be justified by using Nordheim's rules but is easily obtained from this simple modification lends weight to Brennan's proposal.

[9] W. B. Ewbank, L. L. Marino, H. A. Shugart, and H. B. Silsbee, Bull. Am. Phys. Soc. **2**, 383 (1957).

[10] R. M. Elliott and J. Wulff, Phys. Rev. **55**, 170 (1939).

[11] R. L. Christensen, D. R. Hamilton, A. Lemonick, F. M. Pipkin, J. B. Reynolds, and H. H. Stroke, Phys. Rev. **101**, 1389 (1956).

[12] L. W. Nordheim, Phys. Rev. **78**, 294 (1950); Revs. Modern Phys. **23**, 322 (1951).

[13] M. H. Brennan, Bull. Am. Phys. Soc. **5**, 245 (1960) (this reference lists the title and author only).