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Search for Photodeuterium from Copper*

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Copper foil samples were irradiated with bremsstrahlung of 24-, 30-, and 40-MeV peak energy and production of deuterium by the (γ,d) reaction investigated. After irradiation, gas released from the samples was introduced into an omegatron mass spectrometer and the ion currents HHD^+ and HH^+ measured. The sensitivity of the detection was sufficient to use the amount of deuterium in natural hydrogen as calibration. No HHD^+ was observed after irradiation at 24 and 30 MeV and the ratio of deuterium to hydrogen in the irradiated samples was estimated to be less than 0.0016 and 0.00014, respectively. At 40 MeV the ratio was measured to be approximately 0.0009. The ratio of deuterium to hydrogen expected on an evaporation theory has been calculated for several level density formulas. The predicted values are much greater than the observed deuterium to hydrogen ratio. We conclude that deuterons are not evaporated from copper at these photon energies. The direct photointeraction pickup process of Madsen and Henley is estimated to predict a ratio of deuterium to hydrogen of 0.0012 at 40 MeV and 0.0003 at 30 MeV for relatively high residual nucleus excitations. Such predictions seem consistent with the experimental results.

I. INTRODUCTION

EARLY investigation of the interaction of medium-energy electromagnetic radiation with medium-weight nuclei gave indication of the production of anomalously large numbers of deuterons.[1–4] More recent work[5–8] has not confirmed these earlier results. In most of the later work, the reported photodeuteron to photoproton yield ratio lies near the limit of detection of the experimental methods. A more extensive examination of photodeuteron emission, particularly at higher energies, has recently been reported.[9] In order to clear up some of the discrepancies at lower energies, we have investigated the problem with a technique of different type and higher sensitivity.

In this paper we present the results of a photodeuteron investigation using an omegatron mass spectrometer capable of detecting the deuterium present in natural hydrogen gas.

II. APPARATUS

A. Targets

Copper was chosen as the target element because it is one of the elements for which conflicting results had been reported and because of its favorable mechanical properties.

Two copper cups $3\frac{1}{4}$ in. in diameter, 2 in. deep with $\frac{1}{8}$-in.-wall thickness were prepared and into each were inserted 100 pieces of 3-in. copper squares, thickness 0.005 in. Seams on the cups were welded in an inert gas atmosphere without flux. All of this had been acid cleaned and was then attached to the vacuum system by means of a Granville-Phillips type *C* valve.

After rough pumping and baking for several hours at 250°C, the background pressure, as measured on the one liter-per-sec VacIon pump, was of the order of 10^{-8} Torr. The valve to the vacuum pump was then closed off and the target assembly removed from the system. As a further precaution against atmospheric leakage into the target, a squeezed copper tube adapter was used to seal off the high-pressure side of the valve at about 10^{-3} Torr. The target was then ready for exposure.

The adequacy of these targets for revealing photodisintegration products is determined by the diffusion of hydrogen and hydrogen deuteride out of the copper foil and by the permeability of the copper cup to these gases. Calculations based on the diffusion constant reported by Seith[10] for hydrogen in copper, indicate that at room temperature outgassing times of the foils are of the order of 4 min. Measurements of the permeability constant of copper for hydrogen at higher temperatures by Smithels and Ramsey,[11] and by Gorman and Nardella[12] were extrapolated to the temperatures and pressures expected in this experiment. The results indicate that the leakage of hydrogen due

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TABLE I. Details of sample exposures.

Peak bremsstrahlung energy	Exposure time	Total electronic charge to target
24±2 MeV	$2^h\,47^m$	2.42 C
30±2 MeV	$1^h\,52^m$	1.41 C
30±2 MeV	$1^h\,44^m$	0.7 C
34 to 41 MeV	$4^h\,15^m$	4.66 C

to permeation through the cup walls should be less than 1% a year.

B. Exposure

Of several targets prepared as above, one was exposed to the bremsstrahlung beam of the Yale electron accelerator. The other was placed in a position near the first, but outside the direct path of the bremsstrahlung beam. Thus, the neutron environment of both targets was expected to be similar. No deuterium and very little hydrogen was ever detected in the control samples.

Four exposures were made, with details given in Table I.

C. Detector

After irradiation, the sample containers were connected to an omegatron[13] and the gas evolved from the copper foils introduced into the mass spectrometer where the light masses were scanned.

It will be recalled that in an omegatron mass spectrometer with a magnetic field B, the radio-frequency field is set at the cyclotron frequency

$$\omega_c = eB/M\,, \tag{1}$$

where M is the mass of the ions and e is the charge. At this frequency, ions of charge-to-mass ratio e/M will be accelerated in a spiral until they strike the collector electrode. The current reaching this electrode is thus a measure of the pressure of a gas whose ions have the given charge-to-mass ratio.

Ideally, the omegatron is capable of resolving ions whose masses differ by ΔM where

$$M/\Delta M = \omega_c R_0 B / 2E_0\,. \tag{2}$$

R_0 is the distance from the collector electrode to the ionizing stream and E_0 is the peak rf field strength. Combining Eq. (2) with (1) gives the resolution varying as the square of the magnetic field and inversely as the mass of the ion under consideration. The magnetic field was set as high as our equipment would allow: $B = 0.814$ W/m^2. The collection radius $R_0 = 10^{-2}$ m and the field strength $E_0 = 140$ V/m.

The masses to be considered and the ions at these masses are given in Table II. Comparison of the required resolution as shown in Table II (column 3) with that expected from Eq. (2), Table II (column 4) indicates that only the $\mathrm{He}^+ - \mathrm{HHD}^+$ doublet at mass 4 is expected to be resolved with any degree of consistency.

Although ions such as HHH^+ and HHD^+, which are formed by secondary reactions, are not encountered in conventional mass spectrometers at the pressures used in this experiment, the omegatron ion source has a trapping effect on the nonresonant ions. As a result, when tuned to mass 4, the HD^+ ions which are directly produced by the electron beam are trapped in orbits near the electron stream. The secondary reaction $\mathrm{HD}^+ + \mathrm{HH} \rightarrow \mathrm{HHD}^+ + \mathrm{H}$ then occurs. It has been shown that the current due to ions formed in this manner is roughly proportional to the first power of the pressure of the gas which is present[14] and is quite appreciable.

Calibration of the omegatron system was made at a total pressure of 10^{-6} Torr, as measured by the ion pump. The system was calibrated with mixtures of 10%, 1%, and 1.4×10^{-6} deuterium to hydrogen mixtures. A roughly linear relationship between the D/H ratio and the measured currents of HHD^+ and HH^+ ions was observed.

III. RESULTS

The observed yield ratios of deuterons to protons observed in the samples irradiated in the bremsstrahlung beam for the four runs are given in Table III.

In runs I and II, the limit on the omegatron sensitivity was set by the insensitivity of the dc amplifier-type electrometer used as ion current meter. In runs III and IV a more sensitive vibrating reed type of electrometer was used. With this latter detector, the ion current for HHD^+ could be observed with normal hydrogen gas and the 1 in 7000 abundance of D in natural H was used for calibration. In run III no HHD^+ was observed and the conservative lower limit of 0.00014 was based on the easily observed D in normal hydrogen.

These values are appreciably lower than the high ratios originally reported for copper.[1–4] Moreover, they are lower than most of the ratios reported in more recent experiments.[5–9] In view of the rather large un-

TABLE II. Doublet separation and omegatron resolution for pertinent ions.

Mass	Ions	Inverse doublet separation $M/\delta M$	Omegatron resolution $M/\Delta M$
2	HH^+, D^+	1290	1190
3	HHH^+, HD^+	3000	793
4	He^+, HHD^+	145	595

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TABLE III. Observed deuteron to proton ratios for various exposures.

Run	Max. brem. energy	Observed $Y(\gamma,D)/Y(\gamma,p)$
I	24	<0.0016, no HHD$^+$ observed
II	30	<0.0003, no HHD$^+$ observed
III	30	<0.00014, no HHD$^+$ observed
IV	40	$\simeq 0.0009$

certainties associated with the results given in these later experiments, the present values and limits may not be in contradiction.

IV. THEORY

The (gamma, deuteron) reaction may be expected to occur by either of two processes. In the first, the absorbed energy is shared by all of the nucleons or groups of nucleons in the nucleus until a large enough fraction of it becomes concentrated on a preformed deuteron which is then emitted. This concept is based on a compound nucleus model in which evaporation is described in statistical terms. A second photodeuteron process results from the direct interaction of the radiation field either with a preformed deuteron, or with a single nucleon which picks up a partner on leaving the nucleus and emerges as a deuteron.

A. Evaporation

For copper, photon absorption by the nucleus occurs most strongly at the giant resonance peak at about 18 MeV.[15] This energy is shared among the particles in the nucleus. De-excitation of the nucleus by means of neutron, proton, or deuteron emission proceeds according to the probability of evaporating each particle. The deuteron in this picture is considered to be already formed in the nucleus as a point particle of mass 2, spin 1, and charge 1. The capture cross section of a nucleus for such a particle has been calculated by Shapiro[16] and his values with the nuclear radius $R = 1.3\times A^{1/3}$F have been used in our calculations.

It may be shown[17] that a quantity proportional to the probability of evaporating a particle b from a nucleus excited to energy E is

$$(2S+1)M\int_0^{E-E_b} \epsilon\sigma_c(\epsilon)W_R(E-E_b-\epsilon)d\epsilon\,, \qquad (3)$$

where $(2S+1)$ is the spin degeneracy of b, M its mass, $\sigma_c(\epsilon)$ the capture cross section for the inverse (capture) reaction, and $W_R(E-E_b-\epsilon)$ is the level density of the residual nucleus at energy $(E-E_b-\epsilon)$.

Charged particle emission is inhibited by the Coulomb barrier which lowers σ_c at small values of ϵ. The deuteron reaction is further suppressed relative to the proton reaction because of its higher Q value. The binding energies E_b of various particles to the copper nucleus are taken from the Q values listed in Table IV (after Everling *et al.*[18]).

To arrive at quantities to be compared with experiment, expression (3) must be summed over the values of E available from the radiation source and absorbed by the nucleus. When the characteristics of the bremsstrahlung[19] source and of the absorption by the copper nucleus[15] are included, (3) is to be replaced by

$$(2S+1)M\int_0^{\infty} N_\gamma(E)\sigma_{c\gamma}(E) \times\int_0^{E-E_b} \epsilon\sigma_c(\epsilon)W_R(E-E_b-\epsilon)d\epsilon dE\,, \qquad (4)$$

$N_\gamma(E)dE$ being the number of photons in energy interval dE and $\sigma_{c\gamma}(E)$ being the photon capture cross section at this energy.

Evaluation of (4) for the proton and deuteron reaction requires an assumption of the level density of the unstable nickel nuclei of mass 62, 61, 64, and 63. We write the level density as suggested by Cameron[20]

$$W_R=\frac{\exp[9/4+4aU]^{1/2}}{[9/4+4aU]^{1/4}\times[3/2+(9/4+4aU)^{1/2}]^{7/2}} \qquad (5)$$

when U is the excitation energy of the nucleus less a pairing energy of 2.8 MeV for the residual nuclei of the proton reaction and 1.37 MeV for those from the deuteron reaction.

The parameter a in (5) has been estimated from three sources. Blatt and Weisskopf[21] suggest $a=2$ MeV^{-1} based on observation of low lying nuclear levels. Larger values of a are suggested by T. D.

TABLE IV. Nucleon and deuteron binding energies in copper.

Reaction	Q value (MeV)
$\mathrm{Cu}^{-63}(\gamma,n)$	10.84
(γ,p)	6.13
(γ,d)	14.49
$\mathrm{Cu}^{-65}(\gamma,n)$	9.91
(γ,p)	7.45
(γ,d)	14.89

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TABLE V. Values of a (MeV^{-1}) to be used in (5) for calculating level densities.

Suggested level density of	Residual nucleus Ni^{62}	 Ni^{61}	 Ni^{64}	 Ni^{63}
Blatt & Weisskopf[a]	2	2	2	2
Newton[b]	6.1	5.8	6.3	6.2
Thomson[c]	10.1	9.5	10.4	10.2

[a] See Ref. 21. [b] See Ref. 22. [c] See Ref. 23.

Newton's[22] analysis of slow neutron scattering data, and still larger values by D. B. Thomson's[23] work with inelastic neutron scattering at higher energies. Table V gives the values of a used in the calculations.

Evaluation of (4) using these level densities was done on the computer facilities of The National Aviation Facilities Experimental Center, Altantic City, New Jersey.[24] The results of these calculations gives the yield ratios shown in Table VI assuming all preformed deuterons. The values for 24 MeV are consistent with the ratios 0.0003 to 0.0022 calculated by Byerly[1] and the value 0.001 mentioned by Madsen and Henley.[25]

B. The Direct Interaction Theory

Madsen and Henley[25,26] have done a very careful analysis of the photodeuteron effect which may be expected from direct excitation from the nucleus. They find two terms in their evaluation which describe a one-stage and a two-stage pickup mechanism. In the one-stage reaction, the deuteron is lifted as an entity from the nucleus. In the two-stage reaction a nucleon which has absorbed a photon picks up an apposite nucleon and escapes from the nucleus as a deuteron.

The single-stage mechanism is relatively unimportant due to the fact that the effective deuteron charge is multiplied by $(N-Z)/A$. The two-stage pickup mechanism gives a small but significant cross section.

Madsen and Henley's calculations for O^{16} and S^{32} give characteristic excitation curves with peak cross sections of about 100 μb about 5 MeV above an energy which is itself above threshold by the energy of the average triplet spin, triplet isospin levels in the residual nucleus. The cross section falls gradually to about 10% of its peak value some 25 MeV above threshold.

TABLE VI. Deuteron to proton yield ratios using various assumptions concerning level densities under different bremsstrahlung conditions.

Level densities of	Max. brem. energy 24 MeV	 30 MeV	 40 MeV
Blatt & Weisskopf	0.0069	0.090	0.266
Newton	0.0027	0.018	0.062
Thomson	0.00027	0.0046	0.019

A rough estimate of the predictions of this theory for the copper nucleus was arrived at by comparison with the values calculated by Madsen and Henley for S^{32} with account taken of the different numbers of pairs of nucleons available for the formation of a deuteron, i.e., combinations of a neutron and a proton from the top shells giving $S=1$, $T=1$, $T_z=0$.

The number of triplet spin, triplet isospin pairs for copper was taken as 35. The excitation of the states in the residual nucleus which are involved in the process was taken as 3, 6, and 7.5 MeV and the deuteron to proton ratio calculated on the Madsen-Henley theory. The results are shown in Table VII. The higher excitation of the residual states is associated with a lower ratio of dueteron to proton emission.

V. DISCUSSION

Comparing the experimental results in Table III with the predictions of the evaporation models taken from Table VI and summarized in Table VIII, the yield ratio for 30- and 40-MeV bremsstrahlung is much below that expected from such models assuming preformed deuterons. It might be wondered if the evaporated deuteron would be torn apart by the Coulomb field as it emerges and, hence, not yield deuterium.

TABLE VII. Deuteron to proton yield ratios for copper from Madsen-Henley direct pickup process.

Average excitation of states in residual nuclei	Deuteron to proton ratio 30-MeV bremsstrahlung	 40-MeV bremsstrahlung
3 MeV	0.00110	0.0020
6 MeV	0.00049	0.0015
7.5 MeV	0.00028	0.0012

A semiclassical estimate of the probability of electric disintegration can be made by calculating the time-dependent electric field to which the emerging deuteron is subjected assuming a constant deuteron velocity. The Fourier transform of this field gives the frequency distribution of the electromagnetic energy. This "virtual photon spectrum" can be combined with the known photodisintegration cross section of the deuteron to yield the probability P for breakup of the deuteron of energy E_d emitted from the edge of a nucleus of radius R and atomic number Z.

$$P=[FZ^2\alpha/4\pi^2\beta]\times\int_{2.25\,\mathrm{MeV}}^{E_d}[\sigma_d(E_\gamma)/R^2]F^2(a)dE_\gamma/E_\gamma, \quad (6)$$

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where

$$F(a)=\int_0^\infty (1+y)^{-2}\cos ay\,dy\,,\quad a=RE_\gamma/hc\beta\,,$$

β is the deuteron velocity divided by the velocity of of light and σ_d is the known cross section for the photodisintegration of the deuteron as a function of photon energy E_γ. Evaluating this expression for a 6-MeV deuteron emerging from copper gives the probability of breaking apart to be small. This is in agreement with a recent calculation by Gold and Wong.[27] We conclude, then, that the deuteron is not usually available for evaporation and probably does not exist with appreciable probability in the preformed condition in the copper nucleus.

The amount of deuterium observed from copper with 40-MeV bremsstrahlung, giving a deuterium to hydrogen ratio of approximately 0.0009 is not too far from the 0.0012 predicted by the two stage direct interaction for high residual nucleus excitation. An uncertainty of a factor of two probably should be allowed in each number.

TABLE VIII. Comparison of deuteron to proton yield ratios for various evaporation models with observed values.

Maximum bremsstrahlung energy	Theoretical ratios	Observed ratios
24 MeV	0.00027 to 0.0069	<0.0016
30 MeV	0.0046 to 0.090	<0.0014
40 MeV	0.019 to 0.266	~ 0.0009

The experimental upper limit of the ratio at 30 MeV is somewhat lower than the direct interaction picture predicts. However, since this is closer to the thresholds, more uncertainty may accrue to the rough calculations. The experimental results may then be regarded as consistent with the Madsen and Henley theory if the residual nucleus is left in states of relatively high excitation (possibly because of picking up the particle from a closed shell).

This seems equivalent to the conclusions of Chizhov *et al.*[9] that the direct pickup process is verified and that the residual nucleus must be left with sufficient energy to separate a further nucleon.

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Inelastic Proton Scattering from Nuclei with 28 Neutrons*

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The inelastic scattering of 17.45-MeV protons from Ti^{50}, Cr^{52}, Fe^{54}, and V^{51} has been measured. Levels up to 5-MeV excitation were studied and the 30°–90° differential cross sections were measured for most of the levels. Spins and parities were assigned on the basis of the angular distributions and agree well with other experiments. The strength of the various inelastic cross sections were studied using a direct reaction theory with distorted waves. Both the collective model and the shell model of nuclear structure were used to describe the nuclear states. In describing the excitation of the strongly excited levels of the even-A nuclei, the collective model picture yielded a strength parameter, β_l, which agreed within experimental error with the β_l value extracted from Coulomb excitation experiments. The shell-model formulation described this data as well using a two-body Gaussian potential of finite range and depth of 45 MeV. The analysis of V^{51}, however, was better explained using a shell-model analysis.

I. INTRODUCTION

INELASTIC proton scattering has been a useful technique for investigating the level structure of nuclei. For bombarding energies in the range $10<E_p<20$ MeV, the inelastic scattering from nuclei with $A>40$ does not usually depend sensitively on energy, and the direct process as opposed to compound nuclear formation seems to be predominant in exciting low-lying ($\lesssim 5$ MeV) states.[1] Furthermore, recent developments in theoretical techniques[2] have greatly simplified the extraction of nuclear structure information from the experimental data.

The inelastic scattering reaction has been formulated using either collective-model or shell-model wave functions to describe the nuclear states. The collective-model treatment has been very successful in the

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