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Nuclear Resonance Fluorescence in Cu^{65}†

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The 1.114-MeV excited state of Cu^{65} has been investigated by means of nuclear resonant fluorescent scattering and self-absorption experiments. The level width was found to be $(1.51\pm0.38)\times10^{-3}$ eV, corresponding to a mean life of $(4.4\pm1.1)\times10^{-13}$ sec. Angular-distribution measurements of the resonantly scattered gamma rays gave $\delta=(E2/M1)^{1/2}=-0.52_{-0.05}^{+0.07}$. From these values, $B(E2)\downarrow=(0.023\pm0.007)\times10^{-48}e^2\text{cm}^4$ and $B(M1)\downarrow=(0.08\pm0.02)(e\hbar/2Mc)^2$. The lifetime agrees with other recent measurements, but the value for δ disagrees with results from Coulomb-excitation experiments.

I. INTRODUCTION

THE low-lying levels of Cu^{65} have recently been the subject of several investigations. Information about this nucleus is of interest in connection with the core-excitation model of the nucleus. Cline and Heath[1] have summarized the investigations of the decay scheme by use of Ni^{65}; and Coulomb-excitation experiments have been carried out by several groups.[2–4]

In this work we are concerned with properties of the 1.114-MeV excited state of Cu^{65}. Angular-correlation measurements of the gamma rays from the level excited by Coulomb excitation have indicated that this state has a spin and parity of $\frac{5}{2}^-$ and decays by a mixed $E2$-$M1$ transition. The angular-correlation measurements of the 368–1114-keV cascade by Cline and Heath[1] are in agreement with this spin and parity assignment.

The $E2$ partial lifetime can be determined with good accuracy from Coulomb-excitation experiments. This information, together with a knowledge of the $E2/M1$ mixing ratio, enables one to calculate the lifetime of the state. For the 1.114-MeV level, a mean life of less than 10^{-13} sec is obtained with a relatively large error because of the uncertainty in the $E2/M1$ mixing ratio. Another approach is to measure the lifetime of the state directly by use of nuclear resonant fluorescent scattering. In principle, by measuring the resonant self-absorption and angular distribution of the resonantly scattered gamma rays, the total level width and mixing ratio of the $E2$ and $M1$ transitions can be obtained.

According to the decay scheme for Ni^{65}, sufficient energy is available to enable the recoil from transitions populating the 1.114-MeV state of Cu^{65} to compensate for the recoil Doppler shift occurring in the emission and absorption of the 1.114-MeV photon. Therefore, it appeared feasible to investigate the properties of this level by means of nuclear resonance fluorescence.

II. EXPERIMENTAL DETAILS

1. Source

The lifetime was expected to be somewhat longer than the collision times for atoms in a solid or liquid. Hence, it was desirable to use a gaseous source so that only a negligible fraction of the recoiling Cu^{65} nuclei would undergo collisions with a resultant loss of memory before being de-excited to the ground state. For practical reasons, it was necessary to find an easily produced stable nickel compound having a boiling point below 1200°C. We found only one compound that fulfilled these conditions, namely, anhydrous $NiCl_2$ which has a boiling point of 975°C.

The recoil energy imparted to the Cu atom in the decay of Ni^{65} can be as high as 16.5 eV. This energy usually will be shared among the atoms of the $NiCl_2$ molecule and can cause the molecule to break up. Be-

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[1] J. E. Cline and R. L. Heath, Phys. Rev. **131**, 296 (1963).
[2] B. Elbek, H. E. Gove, and B. Herskind (to be published).
[3] R. L. Robinson, F. K. McGowan, and P. H. Stelson (to be published).
[4] K. I. Erokhina and I. Kh. Lemberg, Bull. Acad. Sci. (USSR) Phys. Ser. **26**, 205 (1962).

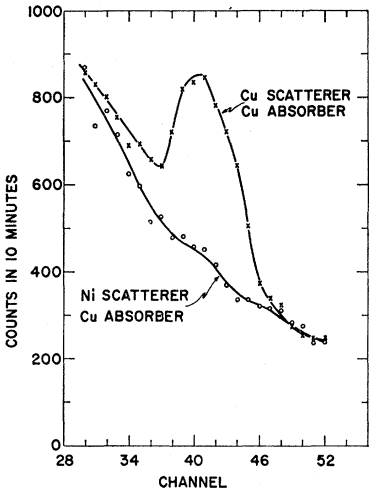

FIG. 1. Resonantly scattered 1.114-MeV quanta. The upper curve shows the scattering with the Cu scatterer and absorber in position and the lower with the comparison Ni scatterer. The runs were of 10-min duration. The nonresonant scattering data were corrected for source decay.

cause the details of the energy sharing processes are complicated and unknown, the effective source strength for photons having the proper energy to be resonantly scattered cannot be determined. Fortunately, this trouble can be avoided by carrying out a resonant self-absorption experiment.

The samples of $NiCl_2$ were prepared by adding concentrated HCl to metallic nickel enriched to 99.5% Ni^{64}. The mixture was gently heated in a quartz ampoule until the nickel disappeared. Then the system was evacuated and heated further until only anhydrous $NiCl_2$ remained. In the first preparations, to help eliminate possible contaminants, the $NiCl_2$ was distilled into a second quartz ampoule which was then sealed off. Later work showed this precaution to be unnecessary. The two strongest sources used in the final runs each contained about 14-mg $NiCl_2$ in 5-cc ampoules.

The encapsulated samples were irradiated for periods of 6–24 h in a neutron flux of $(5–7)\times10^{13}$ neutrons cm^{-2} sec^{-1} in the Argonne CP-5 reactor. The ampoules were then heated to 1025°C to vaporize the source. Tests made with variations of up to $\pm25°$ about this operating temperature showed no change in the resonant fluorescent scattering.

The irradiation of the quartz ampoules produced some undersirable background activities, notably those of Si^{31} and Cl^{38}. The strong Si^{31} activity had a half-life of 2.5 h and decayed over 99.9% by pure β^- emission. Consequently, its background contribution consisted chiefly of bremsstrahlung and caused little trouble. The 37-min Cl^{38} activity, however, contained high-energy gamma rays which after being Compton scattered could still give rise to a varying background in the 1.114-MeV region of interest. This difficulty was avoided by delaying the data taking until after the Cl^{38} activity had decreased by an order of magnitude.

The disadvantage of the undesirable Si^{31} and Cl^{38} activities was more than offset by the ease of handling the relatively strong Ni^{65} activities produced and by the fact that the ampoules could be used several times for the complete cycle of irradiation plus heating with no sign of physical deterioration and negligible buildup of long-lived background activity.

2. Self-Absorption Measurements

A conventional cylindrical symmetry was used for the resonant scattering experiments.[5] The resonant scatterer consisted of a Cu ring, 1 ft in diameter, 4 in. high, and $\frac{1}{2}$ in. thick. A similar one of nickel was used for comparison measurements. A 3×3-in. NaI(Tl) crystal was used to detect the resonantly scattered photons and the data were recorded with a multichannel pulse-height analyzer. No resonant scattering was noted with the source at room temperature in the preliminary runs, although an effect amounting to a few percent above background could have escaped detection.

The self-absorption measurements were made with Cu and Ni absorbers which had been matched to within $\frac{1}{2}$% for electronic absorption at room temperature by use of the 1.114-MeV gamma rays from a Zn^{65} source. The absorbers were lined with Al foil which acted as a heat reflector and served to keep their temperatures within 10°C of room temperature. Each run to determine the resonant absorption was of 10-min duration, and the Ni and Cu absorbers were alternated so that corrections for source decay and a changing dead time in the analyzer were limited to less than 1%. After

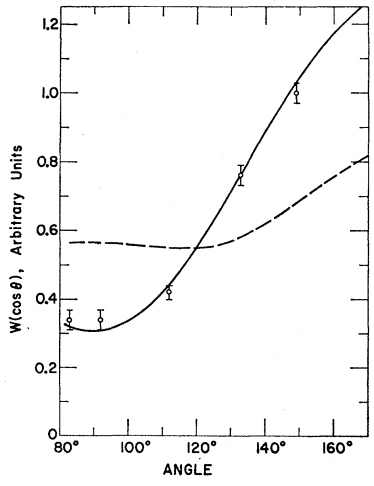

FIG. 2. Angular-correlation data. The solid curve represents the fit of the data for $A_2=1.02$ and $A_4=0.07$. The dashed curve shows the theoretical angular distribution, corrected for the finite angular resolution, for the assumption of pure $E2$ transitions for a spin sequence of $\frac{3}{2}-\frac{7}{2}-\frac{3}{2}$. The two curves are normalized at 120°.

[5] G. B. Beard and W. H. Kelly, Nucl. Phys. 43, 523 (1963).

corrections for source decay were made, individual background contributions measured with the Cu and Ni absorbers and the Ni scatterer agreed to within statistical uncertainties. An average of these was then used for background corrections. Figure 1 shows the observed effect in typical 10-min runs with the Cu absorber in position. As a check, the number of resonantly scattered photons under the photopeak for various runs was plotted against time. The half-lives obtained in this way agreed with the half-life for the decay of Ni^{65}.

3. Angular-Distribution Measurements

For the angular-distribution measurements, the 3-×3-in. NaI(Tl) crystal was replaced by a 2-×2-in. crystal. The Cu scatterer remained fixed and the crystal position was varied to change the scattering angle over the range from about 90° to 150°.

III. EXPERIMENTAL RESULTS

1. Lifetime

The resonant absorption was found by forming the ratio $(C_{Ni}-C_{Cu})/(C_{Ni}-b)$, where C_{Ni} and C_{Cu} are the counting rates for Ni and Cu absorbers, respectively, and b is the background rate. An average of four runs gave an absorption of $(5.6\pm1.4)\%$ for an average path length of 0.33 in. in the copper absorber. As a first approximation,[6] the equation relating the level width Γ to the resonant absorption A is

$$A=nd(g_2/g_1)\Gamma\lambda^2/4[\pi(\Delta_a^2+\Delta_s^2)]^{1/2},$$

where

$$\Delta_\nu^2=2kT_\nu E^2/Mc^2.$$

Here n is the number of resonant scatterers per cm^3, d the average absorber thickness (in cm), g_2/g_1 the statistical factor, λ the wavelength of the gamma of energy E, k the Boltzmann constant, T_ν the effective temperature of the absorber or scatterer, M the atomic mass of the resonant nucleus, c the speed of light, and Δ_a and Δ_s are the Doppler widths of absorber and scatterer, respectively.

The numerical values are as follows: $n=2.61\times10^{22}$ atom/cm^3, $g_2/g_1=\frac{3}{2}$, $\lambda^2=1.24\times10^{-20}$ cm^2, and (by use of a Debye temperature [7]of 330°C and the Lamb correction for crystalline binding[6]) $\Delta_a=1.08$ eV and $\Delta_s=1.06$ eV. This gives a level width of 1.46×10^{-3} eV for an absorption of 5.6%. However, there was a relatively large self-absorption in the $\frac{1}{2}$-in.-thick scatterer. Taking this into account, a more accurate calculation gives a level width of $(1.51\pm0.38)\times10^{-3}$ eV for the 1.114-MeV state of Cu^{65}. From the uncertainty principle, the corresponding mean life is $(4.4\pm1.1)\times10^{-13}$ sec.

[6] F. R. Metzger, Phys. Rev. **103**, 983 (1956); *Progress in Nuclear Physics*, edited by O. R. Frisch (Pergamon Press, London, 1959), Vol. 7, p. 54.

[7] N. F. Mott and H. Jones, *The Theory of the Properties of Metals and Alloys* (Dover Publications, New York, 1958), p. 14.

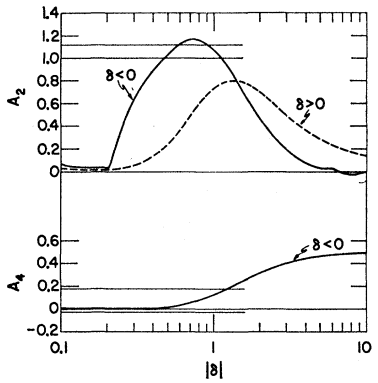

FIG. 3. Theoretical values of A_2 and A_4 as a function of the mixing ratio $\delta=(E2/M1)^{1/2}$. The solid curves represent negative values of δ while the dashed curve for A_2 is for positive δ. The pairs of lines show the experimentally allowed ranges of A_2 and A_4, after correction for the finite angular resolution.

2. Angular Distribution

Figure 2 shows the average angular distribution of two separate runs. The solid curve shows $W(\cos\theta)=1+A_2{}^2P_2(\cos\theta)+A_4{}^2P_4(\cos\theta)$, where $P_2(\cos\theta)$ and $P_4(\cos\theta)$ are Legendre polynomials, $A_2=1.02$, and $A_4=0.07$. These values for A_2 and A_4 were obtained by a least-squares fit to the data points. The dashed line shows the theoretical angular distribution for pure $E2$ transitions for $\frac{3}{2}-\frac{7}{2}-\frac{3}{2}$ spin sequence. The experimental points clearly are in disagreement with a $\frac{7}{2}$ spin assignment to the 1.114 MeV state.

Theoretical values of A_2 and A_4 as a function of the mixing ratio $\delta=(E2/M1)^{1/2}$ are shown in Fig. 3 on the assumption that the decay proceeds by a $\frac{3}{2}-\frac{5}{2}-\frac{3}{2}$ spin sequence with the same $E2/M1$ mixing ratio in each part. The pairs of straight lines show the limits on the values of A_2 and A_4 based on results of the present work. A correction for the finite angular resolution has been made. From the curves it can be seen that either $\delta=-0.52_{-0.05}{}^{+0.07}$ or $\delta=-1.02_{-0.12}{}^{+0.09}$ will give agreement with the experimental results. It will be shown in the discussion that only $\delta=-0.52$ is compatible with other results for the $E2$ and $M1$ partial lifetimes.

IV. DISCUSSION

By combining the measured values of $(4.4\pm1.1)\times10^{-13}$ sec for the mean life and $\delta=-0.52_{-0.05}{}^{+0.07}$ one finds an $E2$ partial lifetime $\tau(E2)=(2.1\pm0.7)\times10^{-12}$ sec and an $M1$ partial lifetime $\tau(M1)=(5.6\pm1.5)\times10^{-13}$ sec. If follows that the reduced transition probabilities for de-excitation are $B(E2)\downarrow=(0.023\pm0.007)\times10^{-48}$ e^2cm^4 and $B(M1)\downarrow=(0.08\pm0.02)(e\hbar/2Mc)^2$. On the other hand, for $\delta=-1.02_{-1.02}{}^{+0.09}$, $\tau(E2)=(8.6\pm2.3)\times10^{-13}$ sec and $\tau(M1)=(9.0\pm2.5)\times10^{-13}$ sec.

Recently, other measurements of the lifetime of the 1.114-MeV state have been made, one by the Doppler-

TABLE I. Values for $B(E2)\downarrow$ and the corresponding $\tau(E2)$ from Coulomb-excitation measurements.

$B(E2)\downarrow$ ($e^2\times10^{-48}$ cm^4)	$\tau(E2)$ (10^{-12} sec)	Reference
0.0170	2.9	Elbek *et al.* (Ref. 2)
0.019	2.6	Erokhina and Lemberg (Ref. 4)
0.0230±0.0025	2.14±0.23	Robinson *et al.* (Ref. 3)
0.018 ±0.003	2.7 ±0.5	Temmer and Heydenburg[a]

[a] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **104**, 967 (1956).

shift method[8] and another using nuclear resonance fluorescence.[9] Tables I and II show the results for these measurements as well as the $B(E2)\downarrow$ and corresponding $E2$ partial lifetimes obtained from Coulomb-excitation measurements. A weighted average[10] of these values for $\tau(E2)$ and τ lead to $\tau(E2)=(2.60\pm0.14)\times10^{-12}$ sec, $\tau(M1)=(6.7\pm0.8)\times10^{-13}$ sec, and $\delta^2=0.26\pm0.03$. Of the two values of the mixing ratio δ that were consistent with the angular distribution coefficient A_2 in Fig. 3, only $\delta=-0.52$ is consistent with this value of δ^2.

With this choice for δ, the values of $\tau(E2)$ and τ obtained from the different experiments are in good agreement. However, there still remains the unexplained discrepancy between the mixing ratios obtained from the angular distributions of gamma rays following Coulomb excitation and the value found in the present work. Elbek, Gove, and Herskind,[2] using oxygen ions for excitation, find $\delta=-0.30\pm0.13$, while Robinson, McGowan, and Stelson[3] and $\delta=-0.22\pm0.06$ with excitation by alpha particles. Such low values of δ are incompatible with the results for $\tau(E2)$ in Coulomb-excitation experiments and the measured lifetime τ for the 1.114-MeV state of Cu^{65}.

The mixing ratio is of particular interest here because the presence of the $M1$ contribution is in direct contradiction to the prediction of the simple core-excitation model. This model pictures the Cu^{65} nucleus as an even-even core with the odd proton in a $p_{3/2}$ orbit coupled to the core. The ground state should therefore be $\frac{3}{2}^-$ with four excited states of spin $\frac{1}{2}^-$, $\frac{3}{2}^-$, $\frac{5}{2}^-$, and $\frac{7}{2}^-$, all of which should decay by $E2$ transitions to the ground state since $M1$ transitions are forbidden. However, at the same time many of the predictions of the core-excitation model appear to be valid for odd-A nuclei. Hence various modifications to the simple model are being considered to account for observed discrepancies such as the high $M1$ admixtures in the deexcitation of states[11] in Cu^{65} and Cu^{63}.

TABLE II. Measured values of the total lifetime τ.

τ (10^{-13} sec)	Method	Reference
$5.3_{-0.04}{}^{+0.05}$	Doppler shift	Eswaran *et al.* (Ref. 8)
4.4±1.1	Resonance fluorescence	This work
6.5±1.6	Resonance fluorescence	Kaipov *et al.* (Ref. 9)

[8] M. A. Eswaran, H. E. Gove, A. E. Litherland, and C. Broude (to be published).

[9] D. K. Kaipov, R. B. Begzhanov, A. V. Kuziminov, and Yu. K. Shubnyi, Zh. Eksperim. i Teor. Fiz. 44, 1811 (1963) [English transl.: Soviet Phys.—JETP **17**, 1217 (1963)].

[10] In taking this average, uncertainties of about 15% were arbitrarily assigned to the $B(E2)$ values which do not have errors assigned in Table 1.

TABLE III. Summary of properties of the 1.114-MeV state. Rows 7 and 8 give the ratio of the observed transition probabilities to the Weisskopf estimate[a] for the $E2$ and $M1$ transitions, respectively. ($R=1.2A^{1/3}\times10^{-13}$ cm.)

$B(E2)\downarrow=(0.019\pm0.001)e^2\times10^{-48}$ cm^4
$\tau(E2)=(2.60\pm0.14)\times10^{-12}$ sec
$B(M1)\downarrow=(0.065\pm0.008)(e\hbar/2Mc)^2$
$\tau(M1)=(6.7\pm0.8)\times10^{-13}$ sec
$\tau=5.3_{-0.4}{}^{+0.5}\times10^{-13}$ sec
$\Gamma=1.25_{-0.12}{}^{+0.09}\times10^{-3}$ eV
$T(E2)/T(E2)_w=12$
$T(M1)/T(M1)_w=0.034$
$\delta=-0.51\pm0.03$

[a] D. H. Wilkinson in *Nuclear Spectroscopy*, edited by F. Ajzenberg-Selove (Academic Press Inc., New York, 1960), p. 858.

Table III summarizes the properties under discussion for the 1.114-MeV level in Cu^{65}. The values shown are based on weighted averages of the results from both Coulomb excitation and resonance-fluorescence experiments, except for the exclusion of the anomalously low values of δ from the Coulomb-excitation angular-distribution measurements.

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[11] For recent discussions in which models based on single-particle coupling to an even-even core are applied to copper isotopes, see H. E. Gove, Phys. Letters 4, 249 (1963); M. Harvey, Nucl. Phys. 48, 578 (1963); Ref. 3; and J. B. Cumming and N. T. Porile, Phys. Rev. **122**, 1267 (1961). See also other work cited in these references.