RESONANCE SCATTERING OF γ QUANTA ON Cu⁶⁵ AND Ti⁴⁶

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The lifetimes of the excited states of Cu⁶⁵ and Ti⁴⁶ nuclei at 1.114 and 0.890 MeV, respectively, are measured by nuclear resonance scattering using gaseous sources of Ni⁶⁵ and Sc⁴⁶ in NiCl₂ and ScCl₃. A lifetime $(1.42 \pm 0.20) \times 10^{-11}$ sec was obtained for the 0.890-MeV level of Ti⁴⁶. The lifetime of the Cu⁶⁵ 1.114-MeV level was found to be $(6.5 \pm 1.6) \times 10^{-13}$ sec, corresponding to $\tau_{\gamma} = 8.3 \times 10^{-13}$ sec for the M1 transition and to the E2/M1 intensity ratio $\delta^2 = 0.32$.

WE have used nuclear resonance scattering to determine the lifetimes of the second excited Cu⁶⁵ state (1.114 MeV) and the first excited state of the even-even nucleus Ti⁴⁶ (0.890 MeV). The sources were Ni⁶⁵ (T_{1/2} = 2.56 hours) and Sc⁴⁶ (T_{1/2} = 80 days).

When the source is an isotope that decays to an excited state of the desired nucleus, some of the transition energy is expended for nuclear recoil and γ quanta are emitted and absorbed. In the present case the energy losses are 20.4 eV for Cu^{65} and 18.4 eV for Ti⁴⁶. Since the excited level widths are greatly exceeded by the recoil losses for resonance excitation, the energy of the emitted γ quanta must be restored to the resonance value. This comes about through the Doppler broadening of the γ lines due to recoils from preceding β and γ transitions. In order to exclude effects due to collisions of the recoil nuclei with nearby atoms of the source material, we used the compounds $NiCl_2$ and $ScCl_3$ with the sublimation points 980° and 800°C, respectively.

DETERMINATION OF EXCITED STATE LIFE-TIMES

The lifetime τ_{γ} of an excited nucleus can be determined from the resonance intensity and by the self-absorption method. The first method is known to give a unique value of τ_{γ} for monatomic sources, and also when molecular compounds are used, if chemical bonds are not broken and the entire molecule undergoes recoil. To determine τ_{γ} in the latter case it is necessary to measure the mean cross section $\bar{\sigma}$ for resonance scattering and to calculate P(E), the relative number of γ quanta per 1-eV interval in the resonance region. The relation between $\bar{\sigma}$ and P(E) is given by the familiar equation

$$\bar{\sigma} = \frac{g_2}{g_1} \frac{\lambda^2}{4} \Gamma_{\gamma} P (E), \qquad (1)$$

where g_2 and g_1 are the statistical weights depending on the spins of the excited and ground states, λ is the resonant wavelength, and Γ_{γ} is the natural level width, which is related to the lifetime by $\tau_{\gamma}\Gamma_{\gamma} = \hbar$ (\hbar is the Planck constant).

The energy distributions of the γ quanta (the microspectra) were calculated from the Ni⁶⁵ and Sc⁴⁶ decay schemes assuming that the recoil nucleus is free and that there are no β - γ correlations. For Ni⁶⁵ we obtained P(E) = 0.0084/eV, taking into account the cascade β (1.010 MeV) $-\beta$ (0.620 MeV) $-\gamma$ (0.370 MeV) leading to the 1.114-MeV level. For Sc⁴⁶ we obtained P(E) = 0.017/eV.

It must be taken into account, however, that when molecular bonds are broken the energy distribution of the γ quanta differs from the monatomic-source case, because a portion of the recoil energy will be used to break chemical bonds, etc. Since this bond-breaking effect cannot be taken into account exactly at the present time, the calculated values of P(E), and therefore of τ_{γ} , can be regarded as upper limits. For Sc⁴⁶, considering the position of the resonance line in the microspectrum, the bond effect can be regarded as insignificant.

To determine the true value of τ_{γ} , the selfabsorption method is used;^[1,2] this consists in investigating the absorption of resonance radiation by a resonant absorber placed between the source and the scatterer. We used this method to determine τ_{γ} of the 1.114-MeV Cu⁶⁵ state. However, the method was ineffective for the 0.890-MeV Ti⁴⁶ state because of the small level width (~ 10⁻⁵ eV) and the small Ti^{46} content (7.95%) in a natural isotope mixture.

If $\Delta = (E_0/c)(2kT/M)^{1/2}$ (where k is the Boltzmann constant and T is the effective temperature) is the Doppler width of a line resulting from thermal motion of the atoms, then if $\Gamma_{\gamma} \ll \Delta$ the resonance scattering cross section is

$$\sigma = \sigma_0 \frac{\sqrt{\pi}}{2} \frac{\Gamma_{\gamma}}{\Delta} \exp\left[-\left(\frac{E-E_0}{\Delta}\right)^2\right], \qquad (2)$$

where $\sigma_0 = (g_2/g_1)(\lambda^2/2\pi)$ is the cross section for exact resonance, E is the energy of the quantum, and E_0 is the exact resonance energy.

If the resonance effect of the scatterer is $\int N(E)\sigma(E)dE$, where N(E)dE is the number of γ quanta in the interval dE, an absorber d cm thick containing n nuclei/cm³ attenuates the resonance effect by a factor

$$K = \int [1 - n\sigma(E) d] N(E) \sigma(E) dE / \int N(E) \sigma(E) dE.$$
 (3)

Using (2) for σ and assuming that N(E) is a smooth function of the energy, we obtain

$$K = 1 - \frac{g_2}{g_1} \frac{\lambda^2 \Gamma_{\gamma} n d}{4\pi^{1/2} \Delta}.$$
 (4)

The effective temperature depends on the ratio of the actual to the Debye temperature and can differ for the absorber and scatterer. In this case a suitable correction must be introduced in the last equation.

EXPERIMENT

The Ni⁶⁵ source was produced by thermalneutron irradiation of NiCl₂, prepared from nickel enriched to 77.8% Ni^{64} . The NiCl₂ was contained in an evacuated sealed quartz ampoule, which was bombarded with a neutron flux of $1.8 \times 10^{13}/\text{cm}^2$ -sec at the reactor of the Institute of Nuclear Physics of the Academy of Sciences of the Uzbek SSR for a period of eight hours; one hour later the ampoule was placed in a nichrome furnace, where it was heated to $\sim 1000^\circ C.$ The $\rm Ni^{65}$ and $\rm Sc^{46}$ activities were ~ 20 mCu. When the quartz ampoules containing NiCl₂ and ScCl₃ were heated to $\sim 1000^{\circ}$ C both sources became gases; this was confirmed experimentally by measuring the distribution of active material in each ampoule before heating and at ~ 1000° C.

The experimental arrangement is shown in Fig. 1. Scattered quanta were detected with a NaI(Tl) crystal of 4-cm diameter and height combined with a FÉU-12B photomultiplier. The energy resolution of the spectrometer was $\sim 10\%$ for the 1.114-MeV line of Zn⁶⁵. In order to re-

FIG. 1. Experimental arrangement. 1-electric furnace, 2-source, 3absorber, 4-scatterer, 5-Pb+Cd filter, 6scintillation spectrometers.



duce the number of low-energy pulses an experimentally determined filter consisting of 4 mm Pb and 1 mm Cd was placed between the scatterer and the crystal. In the work with Sc^{46} the scatterers were pressed $20 \times 20 \times 3.5$ -cm plates of TiO_2 and CaO (for comparison); the mean scattering angle was ~ 108°. For Ni⁶⁵ $26 \times 26 \times 1$ -cm plates of copper and iron (for comparison) were used. Resonance scattering on Cu⁶⁵ was also measured with annular geometry, using two scintillation spectrometers situated symmetrically with respect to the source. The annular scatterers were 30 cm in diameter, 1 cm thick, and 10 or 20 cm high. When single-channel pulse analyzers were used (for Ni⁶⁵) the analyzer window was 5 V wide (the half-width of the 1.114-MeV photopeak).

The measurements were performed as follows. $Sc^{46} \rightarrow Ti^{46}$ decay. Using a source at room temperature, and exchanging the resonant TiO₂ and nonresonant CaO scatterers every hour, we obtained the scattering spectrum in the range 0.54-1.26 MeV with a 100-channel AI-100 pulse-height analyzer. Similar measurements were recorded using a gaseous source.

Resonance was determined by analyzing the spectra from the TiO_2 and CaO scatterers. The position of the 0.890-MeV photopeak was checked each hour; the drift did not exceed 1%.

 $Ni^{65} \rightarrow Cu^{65}$ decay. Resonance was investigated with annular geometry as follows. Using solid NiCl₂ and alternating the copper and iron scatterers every three minutes, we recorded the counting rate as a function of time. Similar measurements were obtained for the gaseous source. In both instances the intensity of the direct beam from the source was measured. In order to allow for decay of the source, for each run we measured the ratio of resonance to the direct beam intensity: $(N_{Cu} - N_{Fe})/N$. Under the given conditions this ratio must obviously remain constant.

A 100-channel pulse-height analyzer was used with the geometry shown in Fig. 1, where the position of the absorbers used in the self-absorption experiments is indicated. The efficiencies of the two absorbers (copper for resonance and iron for nonresonant comparison) were compared by attenuating the direct beam, as well as by measuring the attenuation of resonance in the copper absorber compared with the iron. For this purpose we registered the scattering spectrum from the solid and gaseous sources with copper and iron absorbers and with copper and iron scatterers. The counting starts for the different absorbers were synchronized by means of the scintillation spectrometer 6 in Fig. 1, which continuously registered the direct beam intensity and its time variation.

The direct-beam intensity was also measured by the main spectrometer before an experimental run began. The ratio $\Delta N/N$ was determined for each absorber. The efficiency of the copper absorber in attenuating resonance was determined by comparing the ratios $(\Delta N/N)_{Cu}$ and $(\Delta N/N)_{Fe}$; here $\Delta N = N_{Cu} - N_{Fe}$, where N_{Cu} is the count obtained using the copper scatterer.

RESULTS AND DISCUSSION

 $Ni^{65} \rightarrow Cu^{65}$ decay. Table I gives the measurements of $\Delta N = N_{Cu} - N_{Fe}$ (with annular geometry) and the ratio $\Delta N/N$ for the solid and gaseous sources. The counting rates from the two scatterers were identical in the absence of resonance. The change to a gaseous state of the source was accompanied by enhanced scattering from the copper scatterer compared with the iron scatterer due to resonant γ scattering. This effect comprised ~ 6% of the total counting rate. The higher counting rate under nonresonant conditions resulted from the natural background which passed through the shield and was scattered by objects surrounding the apparatus.

Figure 2 shows the scattering spectrum obtained with the AI-100 analyzer and the geometry shown in Fig. 1. In the absence of resonance the scattering spectra from copper and iron were identical. With the gaseous source the peak close to 40 V resulted from resonance. To determine the FIG. 2. Spectrum of resonance scattering on Cu^{65} using a gaseous source. O - from copper scatter; \bullet from iron scatterer.

mean resonance scattering cross section we performed measurements at 90° and 120° scattering angles, obtaining $\sigma_{90^\circ}/\sigma_{120^\circ} = 1.40 \pm 0.20$.

The correlation function for the given case of a mixed E2+M1 transition can be written in the form $f(\theta) = 1 + A_2P_2(\cos \theta)$, where $A_2 = 0.70$ ± 0.20 . Unfortunately, the character of the dependence of A_2 on δ^2 (δ^2 is the E2/M1 intensity ratio) does not permit a unique decision between the values $\frac{5}{2}^{-}$ and $\frac{3}{2}^{-}$ for the spin and parity of the 1.114-MeV Cu⁶⁵ level. However, the value $\frac{5}{2}^{-}$ is preferred in view of other data.

Spin $\frac{5}{2}$ was obtained in $\begin{bmatrix} 3 \end{bmatrix}$ and also in $\begin{bmatrix} 4 \end{bmatrix}$, where the angular correlation of 0.370- and 1.114-MeV quanta was investigated. The same result follows from the structure of Cu⁶³ and Cu⁶⁵ levels; for the 0.960-MeV level of Cu⁶³, $\frac{5}{2}$ follows from several investigations.

From the data on the angular distribution of scattered γ quanta, taking absorption in the scatterer into account, we obtained the mean resonance scattering cross section $\bar{\sigma} = (1.42 \pm 0.14) \times 10^{-26}$ cm². This result and the calculated value of the microspectrum, P(E) = 0.0084/eV, yielded the upper limit 1.8×10^{-12} sec for the lifetime of the 1.114-MeV Cu⁶⁵ state.

We now give the results obtained by the selfabsorption method. The attenuation of the direct

N _{Cu} /3 min	N _{Fe} /3 min	ΔΝ	$10^4 \Delta N/N$	N _{Cu} /3 min	N _{Fe} /3 min	ΔN	$10^4 \Delta N/N$
474.3 ± 3.6 446 ± 2.3 437 ± 3.3 419.6 ± 3.3	Solid 475 ± 3.6 445 ± 2.3 440 ± 3.2 $421,2\pm3.3$	source -0.7 ± 5.1 $+1.0\pm3.2$ -3.0 ± 4.5 -1.6 ± 4.5	$-0.50\pm3.6 \\ +0.59\pm1.9 \\ -2.17\pm3.2 \\ -0.92\pm2.6 \\ 0.75\pm4.6 \\ -0.75\pm4.6 \\ -0.75$	527 ± 4.2 433 ± 3.6 476 ± 3.3 414 ± 3.2	Gaseou 505.2±4.1 407 ±3.3 456 ±3.2 384 ±3.2	s source 21.8 ± 5.8 26.0 ± 4.5 20 ± 4.5 30 ± 4.5 Mean	$ \begin{array}{r} 10.2 \pm 2.6 \\ 14.9 \pm 2.6 \\ 14.6 \pm 3.3 \\ 15.2 \pm 3.3 \\ \end{array} $

Table I

beam and of resonance by the copper and iron absorbers was:

	Direct beam	Resonance	
Iron absorber	100%	100%	
Copper absorber	(98.0 ± 0.2) %	(90.7 ± 2.1)%	

The copper absorber, which was 14.2 g/cm² thick, reduced resonance $7.4 \pm 2.1\%$ more than the iron absorber.

The actual temperatures of the absorber and scatterer were 320° and 290°C. Using the Debye temperature $T_{\odot} = 343^{\circ}$ C of copper, we obtain the effective temperatures 339° and 311°C, respectively. Using $\lambda^2 = 1.24 \times 10^{-20}$ cm², n = 2.6 $\times 10^{22}$ cm, and $g_2/g_1 = \frac{3}{2}$, we obtain $\Gamma_{\gamma} = 1.01 \times 10^{-3}$ eV, which corresponds to (6.5 ± 1.6) $\times 10^{-13}$ sec for the lifetime of the excited 1.114-MeV Cu⁶⁵ state.

In [5,6] E2 transition probabilities were investigated by means of Coulomb excitation; 2.5×10^{-12} and 2.6×10^{-12} sec were obtained for the lifetime of the 1.114-MeV level. By comparing our value of τ_{γ} with $\tau_{\gamma}(E_2)$ from [5,6], we obtained the lifetime for the M1 transition and consequently the E2/M1 intensity ratio (δ^2) .

For $\tau_{\gamma}(M1)$ we obtain 8.3×10^{-13} sec and $\delta^2 = 0.32$. A comparison of this value with Moszkowski's calculations for one-particle transitions shows that the M1 ground-state transition from 1.114 MeV is hindered by a factor of 40. This disagrees with the model in ^[7], according to which M1 ground-state transitions from excited Cu⁶⁵ states are forbidden. In ^[8] a vibrational model was used to calculate E2 and M1 transition probabilities in odd Cu nuclei. The calculations for Cu⁶³ agreed satisfactorily with experiment.

<u>Sc⁴⁶</u> \rightarrow Ti⁴⁶ decay. The scattering spectrum from a gaseous source was investigated. In the absence of resonance (with solid ScCl₃) identical scattering from the two scatterers was observed. The resonance effect was ~ 8–10%. To determine γ absorption in the scatterer the linear attenuation factor μ was determined experimentally. The solid angle of the scatterer at the source was calculated by integrating over the surface of the scatterer.

Taking the angular distribution of scattered γ quanta for the spin sequence 0-2-0, the mean resonance scattering cross section was calculated to be $(1.91 \pm 0.16) \times 10^{-26}$ cm². Using the foregoing relation (1) between σ and P(E) = 0.017/eV, we obtain $\tau_{\gamma} = (1.42 \pm 0.20) \times 10^{-11}$ sec. As already stated, the value of τ_{γ} will probably not be strongly affected by taking chemical bonds into effect.

The lifetime of the 0.890-MeV level of Ti⁴⁶ was

investigated in ^[9-11] using delayed coincidences; the respective values obtained for τ_{γ} were: < 4 × 10⁻¹¹, < 1.43 × 10⁻¹⁰, and 1 × 10⁻¹² sec. Heydenburg and Temmer^[6] obtained $\tau_{\gamma} = 1.29$ × 10⁻¹¹ sec by the Coulomb excitation method, which was also used in ^[12] to obtain $\tau_{\gamma} = 8.7$ × 10⁻¹² sec.

The Weisskopf equation based on the singleparticle model gives $\tau_{\rm S-p} = 1.45 \times 10^{-10}$ sec. Therefore the 0.890-MeV E2 transition is accelerated by a factor of 10; this indicates the collective character of the excitation.

In the collective model^[13] the lowest excited states of even-even nuclei in the interval $40 \le A \le 150$ are considered to result from quadrupole vibrations of the nuclear surface around an equilibrium spherical shape. In the vibrational model ^[14] the reduced transition probability B(E2, 2→0) is associated with an effective surface tension C₂ and inertial parameter B₂ (characterizing the mass of nuclear matter associated with surface vibrations) by the relation

$$B (E2, 2 \to 0) = \left(\frac{3}{4\pi} ZeR_0^2\right)^2 \frac{\hbar^2}{2 (B_2C_2)^{1/2}};$$

$$E = \hbar (C_2/B_2)^{1/2}, R = 1.2 \cdot 10^{-13} A^{1/3}.$$
 (5)

Using the experimental values for B(E2) and E, we determine the parameters C_2 and B_2 from

$$C_{2} = \frac{0,995 Z^{2}E}{[B (E2)/B (E2) _{s-p}]},$$

$$\frac{B_{2}}{B_{2}hydrod} = \frac{241Z^{2}A^{-1/_{s}}}{E [B (E2)/B (E2) _{s-p}]}.$$
(6)

The values of B₂ are given in units of B_{2-hydrod}, the inertial parameter corresponding to surface vibrations of an irrotational incompressible liquid drop (the hydrodynamical model). From the value of B(E2) for the first excited 2⁺ level we can also calculate the rms vibrational amplitude $\sqrt{\langle \beta^2 \rangle}$, using the relation

$$B (E2) = \left(\frac{3}{4\pi} e Z R_0^2\right) \langle \beta^2 \rangle.$$
 (7)

The parameter $\sqrt{\langle \beta^2 \rangle}$ can be considered characteristic of the effective nuclear deformation due both to zero-point vibrations and to possible deviations of the equilibrium shape from spherical symmetry.

Table II gives the vibrational parameters C_2 , B_2 , and $\sqrt{\langle \beta^2 \rangle}$ for Ti⁴⁶ and Ti⁴⁸ taking into account all data obtained for the corresponding values of B (E2, 2-0). It follows from Table II that for the given range of nuclei we have an empirical law

1221

Nucleus	E_1 , MeV	E ₂ /E ₁	$B(E2,2\rightarrow O) \\ e^{2} \cdot \mathbf{b}^{2}$	$\frac{B(E2,2\rightarrow O)}{B \text{ s-p}}$	C2, MeV	$\frac{B_2}{B_2 \text{ hydrod}}$	$\sqrt{<\beta^2>}$
Ti ⁴⁶ Ti ⁴⁸ Ti ⁵⁾	$0.890 \\ 0.990 \\ 1.54$	$2.26 \\ 2.337 \\ 1.735$	0.0122 0.0107 —	12.2 10.7	35 45 —	18.2 17.4 —	$0.243 \\ 0.231 \\ -$

to the effect that decreasing energy of the lowest collective level is accompanied by an increasing value of B(E2).

It must be mentioned that the observed laws governing the properties of excited states in eveneven nuclei can be accounted for without using the vibrational model. This has been shown by Raz,^[15] who performed the appropriate calculations for two equivalent particles in the $f_{7/2}$ state using both collective and two-body interactions. For nuclei having only a closed shell, Kisslinger and Sorensen^[16] calculated energy levels and γ -transition probabilities on the basis of two-body interactions, obtaining 0.015 e² - b² for B(E2) in the case of Ti⁵⁰. Unfortunately, a lack of data prevented comparison with experiment in this case.

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Note added in proof (April 10, 1963). The transition probability from the 1.54-MeV level of Ti^{50} has been measured by Vasil'eva, Erokhina, and Lemberg [Izv. AN SSSR, Ser. Fiz. 26, 999 (1962), Columbia Tech. Transl. p. 1007] who obtained B(E2, 0 \rightarrow 2) = 0.4 e² - b².

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