impact parameter are those resulting in compound nucleus formation, then the energy and angular momentum balance become reasonable. Under these assumptions the number of particles emitted per compound nucleus formed would be 2.6, 4.0 and 2.7 for α, p, and n, respectively. The $\bar{E}$ of Table I would be increased by a factor of 2, the $\bar{l}$ increased by a factor of 3, the excitation energy to be accounted for remains the same, and the average angular momentum to be accounted for would be reduced by a factor of $\sqrt{2}$.

The remainder of the collision cross section presumably gives rise to direct-interaction products. The cross section for direct interaction is known to be large in the O—Al[20] reaction. Our preliminary results indicate that fragments from Li to F are produced in the O—Ni reaction, that they are sharply peaked in the forward direction, and the cross section for these processes is substantial.

[20] C. E. Anderson, W. J. Knox, A. R. Quinton, and G. R. Bach, Phys. Rev. Letters **3**, 557 (1959).

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Nuclear Resonance Fluorescence in Cu^{63}: Lifetimes of Excited States and the Slowing Down of Recoils (3–100 ev) in Condensed Media*

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Measurements of the lifetimes of the 669-kev and 963-kev states of Cu^{63} have been performed using the nuclear resonance fluorescence technique. Self-absorption experiments yield mean lives of $(2.94\pm0.24)\times10^{-13}$ sec for the spin $\frac{1}{2}$ 669-kev level and $(7.2\pm1.8)\times10^{-13}$ sec for the spin $\frac{5}{2}$ 963-kev level. The resonance yields from sources inside Cu metal and in water solution were measured. The reduction of the resonance yields due to slowing down of the recoils from the β^+ decay was calculated with the use of the model of Vineyard *et al.* for the solid and with an elastic collision model for the liquid environments. These calculations are in good agreement with the measured yields and result in a unique spin assignment to the 669-kev state. The effect of the scatterer temperature on the resonance yield was shown to be in agreement with the "effective temperature" predicted by the theory of Lamb.

I. INTRODUCTION

THE properties of the odd-mass Cu isotopes are particularly interesting since they are composed of a closed shell plus one odd proton in addition to the even number of neutrons. It might therefore be expected that a theoretical treatment of the properties of these nuclei would be relatively simple. Some theoretical investigations of these nuclei have been performed by Lawson and Uretsky.[1]

Extensive experimental investigations of the decay schemes,[2] magnetic moments, and quadrupole moments, have been performed. Coulomb excitation of the lower levels of these nuclei has been observed[3] and extensive inelastic particle scattering experiments have been carried out.

The success of any detailed theoretical study will depend upon the ability of the proposed model to predict the transition probabilities between the low-lying states. The partial lifetimes for the $E2$ part of the transition from the two lowest excited states in Cu^{63} and Cu^{65} are known from Coulomb excitation.[3] It might be expected that there would be substantial $M1$ contributions to these transitions. In 1954, Ilakovac[4] observed the nuclear resonance fluorescence of the 963-kev state in Cu^{63}. His results indicated that this transition is predominantly $M1$.

The improvements in the use of the resonance fluorescence technique suggested that further observations of the resonance in Cu^{63} would provide more accurate data on the lifetime and the yields from condensed sources. In addition to the detection of the 963-kev resonance, a surprisingly large fluorescence yield of the 669-kev first excited state of Cu^{63} was also observed.

Using the self-absorption method it was possible to obtain reliable values of the lifetimes of the 669- and 963-kev levels.[5] During the course of this investigation these resonances and their angular correlations were also observed by Rothem, Metzger, and Swann.[6] Observation of these resonances using a bremsstrahlung source has also been reported by Booth.[7]

Since the resonance yields in this case are large, and since very intense sources (~2 curies) of the parent

* This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] R. D. Lawson and J. L. Uretsky, Phys. Rev. **108**, 1300 (1957).

[2] *Nuclear Data Sheets* (National Academy of Sciences, National Research Council, Washington, D. C.)

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

[4] K. Ilakovac, Proc. Phys. Soc. (London) **A67**, 601 (1954).

[5] A. Sunyar, J. B. Cumming, L. Grodzins, N. T. Porile, and A. Schwarzschild, Bull. Am. Phys. Soc. **4**, 57 (1959).

[6] T. Rothem, E. R. Metzger, and C. P. Swann, Bull. Am. Phys. Soc. **5**, 266 (1960).

[7] E. C. Booth, Bull. Am. Phys. Soc. **5**, 239 (1960).

activity Zn^{63} could be produced, it was possible to study the effects of source environment and scatterer temperature on resonance yields. These studies are particularly interesting for the understanding and interpretation of the resonance fluorescence technique and its applicability to the investigation of some solid-state problems. For both Cu resonances, a Doppler shift of the emitted γ ray by the recoil from the preceding radiation provides compensation for the loss of recoil energy on emission and absorption of the resonance γ ray. From condensed phase sources the resonance yields depend upon details of the slowing down process of recoils from the β decay, and the lifetime of the gamma emitting state. Therefore, the knowledge of the resonance absorption cross section, the recoil stopping behavior, and the resonance yield for the condensed phase source enables one to determine the transition probabilities for both excitation and decay of the state. By application of the principle of detailed balance, the spin of the excited state can be determined in terms of the known ground-state spin. Very little experimental information is available on the stopping of recoils in condensed phases for the energy region (3–100 ev) pertinent to many nuclear resonance fluorescence experiments. This paper presents a detailed calculation of the attenuation of the Cu^{63} resonance expected from metallic Cu and aqueous sources. The calculations for the metallic source are based on the theoretical studies of Cu recoils in Cu by Vineyard *et al.*[8] A simple model is proposed for the slowing down process in water. These models are shown to account very well for the observed attenuations.

II. Zn^{63} DECAY

A partial decay scheme of Zn^{63} is shown in Fig. 1. This scheme shows all the feeding of the 669- and 963-kev levels observed in the decay studies of Cumming and Porile.[9] The indicated electron capture to positron branching ratios are theoretically deduced from the K/β^+ ratio tables of Perlman and Wolfsberg.[10] It has also been assumed that the L/K ratio is $\sim 10\%$ for these transitions. These theoretical values are in agreement with the experimental observations of Cumming and Porile.[9] This decay scheme differs somewhat from that reported by Ricci, Girgis, and Van Lieshout.[11] Their results indicate some feeding of the 669-kev state by γ rays from higher levels, as well as different branching ratios for the β decays.[12]

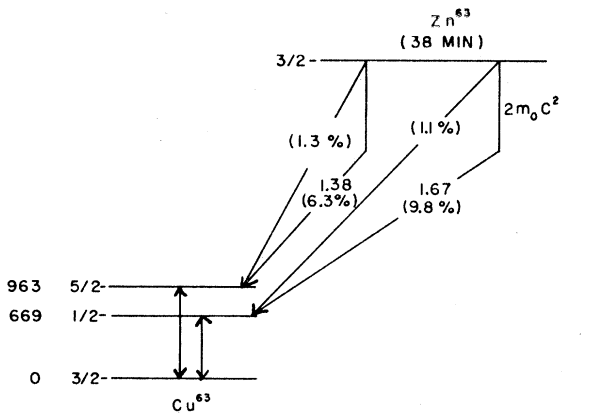

FIG. 1. Partial decay scheme of Zn^{63} showing all the significant feeding of the 669- and 963-kev levels. The main branch of the decay ($\sim 80\%$) is to the ground state of Cu^{63}, and less than 2% of the decays feed levels higher than 963 kev.

The $\frac{3}{2}$ spin of the Cu^{63} ground state has been directly measured. The $\frac{1}{2}$ spin assignment to the 669-kev state is based on the following arguments: If it is assumed that the 770-kev state in Cu^{65} is analogous to the 669-kev state in Cu^{63}, then the absence of β branching to the 770-kev state in the decays of Ni^{65} and Zn^{65}, both with ground-state spins $\frac{5}{2}-$, suggests the spin $\frac{1}{2}$ for these levels. The angular correlations from the 669-kev state observed in Coulomb excitation[13] and in resonance fluorescence[6] are isotropic within experimental errors. For the 669-kev transition, the $E2/M1$ mixing ratio is ~ 0.01 as deduced below from the ratio of Coulomb excitation to resonance fluorescence cross sections. Unfortunately, for this mixing ratio both these angular correlations should be isotropic within several percent for spin assignments of $\frac{3}{2}$ or $\frac{5}{2}$. A spin of $\frac{1}{2}$ necessarily gives an isotropic correlation. It will be shown in Sec. VC that the resonance attenuation due to recoil stopping in solid Cu when combined with the resonance cross section, is consistent only with a spin $\frac{1}{2}$ assignment for this state. This is the only evidence for the $\frac{1}{2}$ assignment which does not depend upon considerations of nuclear systematics.

The $\frac{5}{2}$ spin assignment for the 963-kev state is uniquely determined by the Coulomb excitation angular correlation[13] which is peaked at 90°. (An isotropic angular distribution must result in the Coulomb excitation if the spin were $\frac{3}{2}$, independent of the $E2-M1$ mixing ratio since the correlation factors $F_2(2,\frac{3}{2},\frac{3}{2})$ and $F_4(2,\frac{3}{2},\frac{3}{2})$ as given by Frauenfelder[14] are identically zero.) The $\frac{5}{2}$ assignment is also consistent with the resonance fluorescence correlation[6] as well as with our resonance attenuation measurements.

[8] G. H. Vineyard, J. B. Gibson, A. N. Goland, and M. Milgram, Bull. Am. Phys. Soc. **5**, 26 (1960); J. B. Gibson, A. N. Goland, M. Milgram, and G. H. Vineyard, Phys. Rev. (to be published).

[9] J. B. Cumming and N. T. Porile, Bull. Am. Phys. Soc. **4**, 56 (1959).

[10] M. Perlman and M. Wolfsberg, Brookhaven National Laboratory Report BNL-485, 1958 (unpublished).

[11] R. A. Ricci, R. K. Girgis, and R. van Lieshout, Nuovo cimento **11**, 156 (1959).

[12] Since our absolute resonance yield measurements are performed relative to the individual gamma-ray intensities the disagreement in β^+ branching ratios has no effect in our measurements. The effect of possible gamma-ray feeding of the resonance from higher levels is expected to be small.

[13] F. K. McGowan (private communication).

[14] H. Frauenfelder in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955).

III. EXPERIMENTAL PROCEDURE

The 38-minute Zn^{63} activity was prepared by 10-Mev proton bombardment of a thick Cu target in the Brookhaven 60-in. cyclotron. Except for the measurements of the resonance yields in the solid source, the Cu target was dissolved in HNO_3. The concentration of Cu in the solution along the outgoing beam direction was sufficiently low so that resonant absorption in the source could be neglected in the analysis.

The arrangement of source, scatterer, absorbers and detector is shown in Fig. 2. The scatterer was a 3-in. ×3-in. Cu block which had a surface density of ~13 g/cm² in the incident beam direction. Comparison scatterers of metallic Ni were used in place of the Cu to determine the nonresonant contribution to the spectrum observed in the NaI spectrometer. The pulse-height distribution of the 3-in. ×3-in. NaI(Tl) detector was analyzed with a PENCO 100-channel analyzer.

The most accurate measurements of the resonant self-absorption cross section for both the 669- and 963-kev resonances were obtained using matched Ni and Cu absorbers of a thickness of ~17.0 g/cm² in the beam direction. The Ni and Cu absorbers were matched to better than 2% for the nonresonant transmission at both 669 and 963 kev. Four sets of spectra were measured using the geometry of Fig. 2. These were (a) Ni absorber-Cu scatterer, (b) Ni absorber-Ni scatterer, (c) Cu absorber-Cu scatterer, (d) Cu absorber-Ni scatterer. As expected, the spectra *b* and *d* were identical within statistics in the region of the resonance lines. The spectra *a* and *b* and their difference are shown in Fig. 3. The resonance lines at 669 kev and 963 kev are obvious in the figure and well resolved from the background. The strong 511-kev peak observed in the spectra was shown to be due to annihilation of the positrons created mainly in the scatterer by pair production by the high-energy gamma rays (bremsstrahlung and annihilation in flight of positrons) from the source.

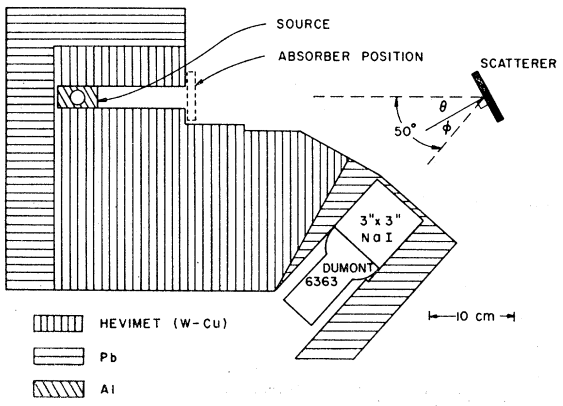

FIG. 2. Experimental arrangement used for observation of the resonance fluorescence. Shielding on top and bottom of the collimator and counter are omitted from the figure.

It is clear that the resonant transmission, T_{res}, of each of the lines is given by the formula

$$T_{res}=(c-d)\epsilon/(a-b), \quad (1)$$

where *a*, *b*, *c*, and *d* are the counting rates in the regions of the photopeaks of the observed resonance lines for the spectra *a*, *b*, *c*, *d* indicated above. ϵ is a number close to 1 which takes account of the slight difference in the electronic absorption and thicknesses for the Cu and Ni absorbers. ϵ was taken as 1.017 on the basis of the calculated electronic absorptions of the Cu and Ni absorbers.

The observed transmissions, T_{res}, were 0.54±0.01 and 0.77±0.05 for the 669-kev and 963-kev resonances, respectively. The widths of the states are derived from these values of T_{res} as shown below.

In order to further check the value of the resonance absorption cross section for the 669-kev transition, as well as to confirm the expected behavior of the absorption with the temperature of the scatterer and absorber, the following experiments were performed:

1. The resonant scattering yields were measured with the scatterer at room temperature (300°K) and at liquid nitrogen (78°K) temperature. In order to maintain the scatterer at liquid nitrogen temperature, it was placed in a styrofoam vessel filled with liquid nitrogen and constructed so that a negligible amount of the coolant was between the source and scatterer and

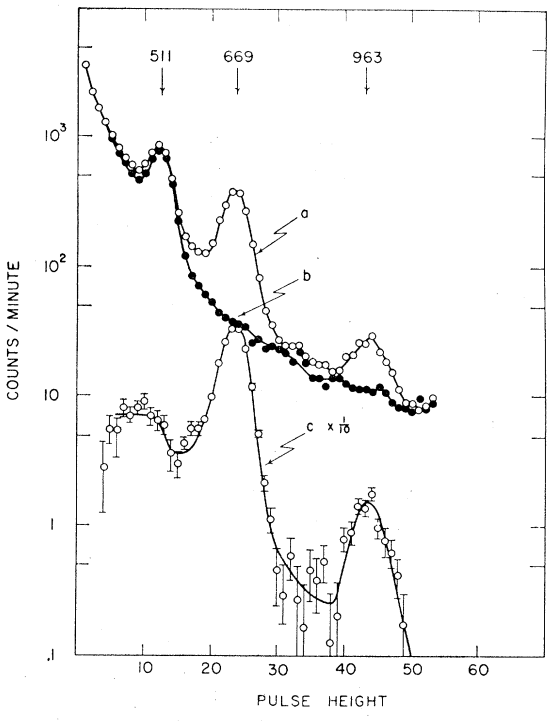

FIG. 3. NaI pulse-height spectrum of the resonances, (a) obtained with a Ni absorber and Cu scatterer. (b) obtained with Ni absorber or Cu absorber and Ni scatterer. The difference between spectra (a) and (b) is shown.

TABLE I. Results of absorption measurements.

Excited level energy (kev)	$\mu_{eff}-\mu_1=\delta k$ (cm^2/g Cu)	k (cm^2/g Cu)	K (barns per atom Cu^{63})	Assumed value of G_2/G_1	Width of state (ev)	Mean life (sec)
669	0.037±0.003	0.055±0.005	8.4±0.7	2/4	2.27×10^{-3}	$(2.94\pm0.24)\times10^{-13}$
963	0.016±0.004	0.023±0.005	3.4±0.8	6/4	9.3×10^{-3}	$(7.2\ \pm1.8)\times10^{-13}$

between the scatterer and counter. A value of 1.095±0.020 was obtained for the ratio of hot (300°K) to cold (78°K) scattering yields.

2. An absorption experiment was performed on the 669-kev resonance with both the scatterer and absorber at liquid nitrogen temperature. The resonant absorption cross section was found to be significantly larger than for the room temperature experiment. With an 11.3 g/cm² Cu absorber, a resonant transmission of 0.55±0.05 was observed.

3. The resonance yield was determined for the resonances as a function of the environment of the Zn^{63} source. Three separate environments were used. They were (1) the solid Cu metal cyclotron target, (2) the Cu metal target dissolved in a solution of HNO_3, and (3) the dissolved source at liquid nitrogen temperature, i.e., frozen. The ratio of the yields obtained from liquid to solid environments was 5.4±0.3 for the 669-kev resonance and 3.5±0.7 for the 963-kev resonance. Within experimental error (10%), the yield for the 669-kev resonance from the frozen source was identical to that from the solution source.

The absolute resonance yields from a liquid source for the 963-kev and 669-kev resonance were determined by ascertaining the source strength by observing the source at a distance of 3 meters with the same NaI detector as was used for the resonance experiment. For the resonant yield measurements a smaller and thinner scatterer was used to simplify the geometric calculations and to minimize the "thick" scatterer effects.

IV. ANALYSIS OF RESULTS

A. Theory

The general theory for the analysis of resonance fluorescence experiments has been presented in the works of Metzger.[15] The analysis of resonance absorption experiments for the particular case of moderately thick absorbers and scatterers and for resonances where the "Doppler width" is large compared to the nuclear level width is given in detail by Ofer and Schwarzschild.[16] In the Appendix of reference 16 it is shown that the yield of resonant gamma rays observed (aside from γ-γ correlation and geometric factors) is given by

$$Y=\frac{\pi^{\frac{1}{2}}N(E_r)\Delta k e^{-(\mu_1+\delta k)a}\left[1-e^{-(\mu_1+\mu_2\cos\theta/\cos\phi+\delta k)b\cos\theta}\right]}{\mu_1+\mu_2\cos\theta/\cos\phi+\delta k}, \quad (2)$$

and that the effective self-absorption for the resonance is characterized by an effective mass absorption coefficient

$$\mu_{eff}=\mu_1+\delta k. \quad (3)$$

The quantities appearing in these formula are defined as follows: $N(E_r)$ is the number of γ rays emitted by the source per unit energy interval at the resonant energy E_r, $\Delta=E_r(2kT/Mc^2)^{\frac{1}{2}}$ is the thermal Doppler width of the absorption line at the effective temperature T, $k\equiv nG_2\lambda^2\Gamma/4G_1\pi^{\frac{1}{2}}\Delta$ where G_2 and G_1 are the statistical weights of the excited and ground states, respectively, λ is the wavelength of the γ radiation, Γ is the natural width of the excited state, and n is the number of nuclei of the scattering isotope per gram of scattering material. μ_1 is the total nonresonant absorption cross section of the scattering material. μ_2 is the poor geometry nonresonant absorption cross section which we have taken as 90% of μ_1 for the analysis of this experiment. The angles θ and ϕ are as shown in Fig. 2. a and b are the thicknesses of the absorber and scatterer, respectively. The quantity δ depends slightly upon the thickness of the absorber and scatterers and the value of k. In all of the analysis of this experiment, δ lies between 0.67 and 0.71. The quantity δ varies because of the change of the emitted line shape in the region of the resonance as the radiation passes through a resonant absorber. The particular value to be used for δ is determined by a procedure of successive approximations to the value of k with the use of Table II of reference 16.

B. Self-Absorption Measurements

The measurement of the resonance transmission using matched absorbers yields essentially the value of $(\mu_{eff}-\mu_1)$. Inserting the values of T_{res} obtained for the 669-kev and 963-kev resonances into the formula $T_{res}=\exp[-(\mu_{eff}-\mu_1)a]$, one obtains the values $\mu_{eff}-\mu_1$ as given in Table I. Considerations of the approximate values of k obtained by setting $\delta=0.70$ and the total thickness of Cu absorber and scatterers (~30 g/cm²) lead to the values of $\delta=0.67$ and $\delta=0.69$ for the 669- and 963-kev resonances, which were then used to compute k as given in Table I. The quantity K is identical to k

[15] F. R. Metzger, in *Progress in Nuclear Physics*, edited by O. R. Frisch (Pergamon Press, London, 1959), Vol. 7; also Phys. Rev. **110**, 123 (1958).

[16] S. Ofer and A. Schwarzschild, Phys. Rev. **116**, 725 (1959).

TABLE II. Hot-cold yield ratios.

Experimental	1.095±0.020
Theoretical	
Lamb eff. temp.	1.088±0.004[a]
Real temp.	1.156±0.012[a]

[a] The quoted errors reflect the error in the value of k measured at room temperature.

except for a change in units. The spins are assumed to be $\frac{3}{2}$ for the ground state, $\frac{1}{2}$ for the 669-kev state, and $\frac{5}{2}$ for the 963-kev state, leading to the values of G_2/G_1 given in the table. The Debye temperature of metallic Cu is 343°K, which yields a ratio of 1.07 for the effective temperature to the real temperature (300°K) according to the theory of Lamb.[17] Using this ratio we calculate values of the width and mean life of the 669- and 963-kev states of Cu^{63} as given in Table I.

The errors in Table I for the 963-kev resonance are purely statistical. It is felt that systematic errors are significantly less than the 25% statistical errors. The statistical errors involved in the measurement of the 669-kev resonance absorption result in only a 4% error in k and reproduce well in different runs. However, this error has been increased by a factor of 2 to account for possible systematic errors.

C. Ratio of Yields for Hot and Cold Scatterers

From the value of the 669-kev resonance cross section given in Table I for 300°K, we have calculated two possible values of k, which may apply when the scatterer is at liquid nitrogen temperature (78°K). One value is obtained from the theory of Lamb which gives an effective temperature of 140°K; the other value results from using the real temperature (78°K). From these values of k, we calculate the expected ratios of hot to cold scattering yields for the thick scatterer. These possible ratios are shown together with the measured ratio in Table II. It is clear that the experimental value agrees with that predicted by the theory of Lamb.[17]

D. Absolute Yield

It is clear from Eq. (2) that the resonance yield from a given scatterer (apart from geometrical factors and the resonance angular correlation) depends only upon k, Δ, and $N(E_r)$. Since Δ is easily calculated and the self-absorption measurements determine the value of k, we can determine the quantity $N(E_r)$ from the absolute yield measurement (if a particular angular correlation is assumed). Table III shows the values of $N(E_r)$ for both the 669- and 963-kev resonances as calculated from the observed resonance yields for the various environments used. In calculating $N(E_r)$ from the experimental data, we have corrected for the angular correlations as measured by Rothem, Metzger, and Swann.[6] There is no angular correlation for the 669-kev resonance. Although the coefficient of $P_2(\cos\theta)$ for the 963-kev resonance was reported as (0.8±0.2), our mean scattering angle of ~130° resulted in a correction to the value of $N(E_r)$ of only 12%.

[17] W. E. Lamb, Phys. Rev. 55, 190 (1939).

V. RESONANCE ATTENUATION IN CONDENSED PHASES

A. General Theory

The calculation of the absolute resonance yield for a source requires knowledge of $N(E_r)$, the number of gamma rays per ev at the resonance energy that are emitted by the source. This may be obtained from a knowledge of the spectrum of recoils of the radiation preceding the resonant γ ray. For the resonances measured in this work, the shape of the emission line is determined primarily by the velocity distribution of the recoils as a result of beta decay and electron capture. However, when the source is in a condensed phase, the recoil velocity distribution and the shape of the emission line are markedly perturbed by collisions of the recoils before gamma-ray emission.

If the velocity of recoils in the condensed phase as a function of time is known, it is possible to calculate the emitted γ-ray spectrum in the region of the resonance. In most cases the γ-ray distribution is continuous over the Doppler broadened resonance and therefore the value of $N(E)$ may be evaluated at a specific energy, $E_r=E_\gamma+E_\gamma^2/Mc^2$, as though the resonance were a δ function.

The probability that a recoil of velocity $v>E_\gamma/Mc$ emit a resonant γ ray is equal to $(c/2E_\gamma v)dE$ (assuming there is no angular correlation between the γ ray and the recoil).[18] If $v<E_\gamma/Mc$ this probability is zero. The slowing down of the recoils may be characterized by the function $v(t,\mathbf{v}_0)$ giving the speed at time t of the recoil which had an initial velocity $\mathbf{v}_0$. The scalar function v may depend on the orientation of $\mathbf{v}_0$ with respect to the environment axes.

$N(E_r)$ for any source may be written as a double integral over the stopping times and initial velocities

$$N(E_r)=\frac{c\int_{v\min}^{v\max}P(v_0)\int_0^T\frac{\exp(-t/\tau)dt}{\tau v(t,\mathbf{v}_0)}d\mathbf{v}_0}{2E_\gamma\int_0^{v\max}P(v_0)d\mathbf{v}_0}. \quad (4)$$

Here T is the time necessary to reduce the velocity of a recoil from v_0 to $v_{\min}=E_\gamma/Mc$. τ is the mean life of the state, and $P(v_0)$ the distribution function of the initial velocities. Where no slowing down or stopping takes place, i.e., a dilute gas source, the time integral in (4) is evaluated from zero to infinity and $v(t,\mathbf{v}_0)$ is equal to

[18] In the following discussion the scalar magnitude $\mathbf{v}\cdot\mathbf{v}/|\mathbf{v}|$ is to be understood for the symbol v.

TABLE III. $N(E_r)$ and attenuations.

Excited level energy (kev)	$N(E_r)$ per gamma ray per 100 ev: Calculated from observed resonance yield, Solid source	Calculated from observed resonance yield, Liquid source	Theoretical[a], Gas source	Attenuations $=A$, Solid source	Attenuations $=A$, Liquid source
669	0.227±0.026	1.22±0.13	2.88	0.079±0.009	0.42 ±0.05
963	0.039±0.009	0.14±0.03	1.65	0.024±0.006	0.085±0.018

[a] Calculated using the decay scheme of Fig. 1. For the positron recoil contribution see footnote 19.

v_0 so that

$$N_{\text{gas}}(E_r)=c\int_{v\min}^{v\max}\frac{P(v_0)}{v_0}dv_0\Big/2E_\gamma\int_0^{v\max}P(v_0)dv_0. \quad (5)$$

We now define the differential attenuation, $\alpha(\mathbf{v}_0)$, which is the attenuation of that part of the gaseous source resonance yield which arises from recoils with initial velocity $\mathbf{v}_0$ as

$$\alpha(\mathbf{v}_0)\equiv\frac{v_0}{\tau}\int_0^T\frac{\exp(-t/\tau)}{v(t,\mathbf{v}_0)}dt. \quad (6)$$

The over-all attenuation of the resonance may be written

$$A\equiv\frac{\int_{v\min}^{v\max}\frac{P(v_0)}{v_0}\alpha(\mathbf{v}_0)dv_0}{\int_{v\min}^{v\max}\frac{P(v_0)}{v_0}dv_0}=\frac{N(E_r)}{N_{\text{gas}}(E_r)}. \quad (7)$$

We shall make certain approximations in the evaluation of the integrals over the initial velocity distribution $P(v_0)$ in the calculation of A. Errors in these approximations will largely be cancelled by the normalization integral in the denominator of Eq. (7). On the other hand, essentially exact calculations of $N_{\text{gas}}(E_r)$ may be obtained from the formulas of Morita *et al.*,[19] and these may be used to calculate values of A from experimentally determined values of $N(E_r)$.

To evaluate the resonance yield from a condensed phase source, $\alpha(\mathbf{v}_0)$ must be obtained for all velocities of interest. In practice this is difficult and simple models are commonly used. For example, Ilakovac[4] assumed copper recoils in water were effectively free for 6×10^{-14} sec (i.e., $v(t,\mathbf{v}_0)=v_0$ for $t<6\times10^{-14}$ sec; $=0$ for $t>6\times10^{-14}$ sec) in analyzing the 963-kev resonance yield from a liquid source. The yield calculated from this model is in agreement with that observed in the present experiment but the agreement must be considered partly fortuitous since the model can only qualitatively be justified because it ignores all details of the slowing down process. Using a similar model Ofer and Schwarzschild[20] have calculated "free times" from measured resonance attenuations of several different nuclear resonances in various environments. They obtained approximately equal magnitudes (within a factor of 3) for the "free times" for the different environments.

B. Resonances Yield from Solid Copper Sources

In an extensive theoretical study, Vineyard *et al.*[8] have studied the motion of copper atoms in a copper crystal. Their numerical calculations have been performed using a high-speed digital computer. The interatomic forces in this model were derived from elasticity and compressibility data and from Fermi-Thomas model considerations. Except for boundary forces, the interatomic potentials in their model are of the form $V=ce^{-r/a}$. Their solutions of the equations of recoil motion were performed in a classical calculation using this potential. Although the model has been used primarily to investigate radiation damage, the results may be used equally well to calculate the resonance attenuation, $\alpha(\mathbf{v}_0)$. Since the sources of Zn^{63} used in the present experiment were produced by bombardment of copper with 10-Mev protons (100 μa for 1 hr) they are subject to extensive radiation damage. However, the bulk of the damage probably anneals rapidly at the temperature of the irradiation (50–100°C). It was therefore assumed that the Zn^{63} atoms occupy reasonably undisturbed lattice sites at the time of the resonance yield measurements. Since the electronic relaxation times in the Cu lattice are much shorter than T it was assumed that the Cu recoils from the β decay of Zn^{63} have equilibrium charge distributions. Application of the computed trajectory calculations thus appears to be justified for the determination of $\alpha(\mathbf{v}_0)$.

[19] M. Morita, R. S. Morita, and M. Yamada, Phys. Rev. **111**, 237 (1958). The formulas in this reference do not give explicit forms for the calculation of $N(E_r)$. Also, some numerical factors have been omitted therein. It can be shown that $N(E_r)$ for a simple β-γ cascade can be determined from the form for $P(\tau,E_0,K)$ given in Eq. (3) of this reference. Using the notation of Morita *et al.* we obtain $P(K)=[P(1,E_0,K)+P(-1,E_0,K)]/4f(\Sigma\ (j_2\|L\|j_1)^2$, where it is assumed that there is no Fierz interference in the β decay, i.e., $b=0$, and $f\equiv\int_1^{E_0}F(Z,E)E(E^2-1)^{\frac{1}{2}}(E_0-E)^2dE$. Explicit integration of the function f for $F(Z,E)=1$ can be found in the work of S. A. Moszkowski, Phys. Rev. **82**, 35 (1951). It is obvious that $N(E_r)=[P(K)][dK/dE_r]=[M/E_r]P(K)$ where M is the recoil mass in units of the electron mass. For both Cu resonance we have assumed that the β decay is predominantly Gamow-Teller and that the recoil-γ angular correlation term may be neglected. It is interesting to note that for both resonances the approximation $F(Z,E)=1$ in the equation for $P(K)$ produces an error of less than 2%. This approximation may be quite poor in other cases if K is close to (E_0-1).

[20] S. Ofer and A. Schwarzschild, Phys. Rev. Letters **3**, 384 (1959).

The output of the computer program describes the motions of all atoms in the crystal as a function of time. If a single atom is given an initial velocity, $\mathbf{v}_0$, then the output for this atom is a table of the velocity vs time necessary for our calculation of $\alpha(\mathbf{v}_0)$. Results for initial recoil energies of 4, 10, 25, 50, and 100 ev were available. The copper crystal is face-centered cubic and its properties should vary with the initial direction of motion. Computer time limitations precluded obtaining data for many directions. However, initial velocities oriented along the three principal directions were investigated; the ⟨100⟩ direction along a cube edge; the ⟨110⟩ direction along a face diagonal; and the ⟨111⟩ direction along a body diagonal. In addition, data were available for several intermediate angles at 25 ev.

Slowing-down curves for several recoil energies with initial velocities in the ⟨100⟩ direction are presented in Fig. 4. The range of recoil velocities of interest lies between 3.4×10^5 cm/sec (3.8 ev), the minimum necessary to emit a 669-kev resonant gamma ray, and 13.8×10^5 cm/sec (62 ev), the recoil velocity due to electron capture proceeding the same transition. The range of velocities for the 963-kev transition lies within these values. It may be noted from Fig. 4 that the time to drop below $v_{\min}$ varies considerably with the initial energy and, even for recoils in a given direction, neither constant time nor constant distance approximation for stopping are justified. There are also variations of the order of a factor of two in the time to drop below resonance for different initial directions

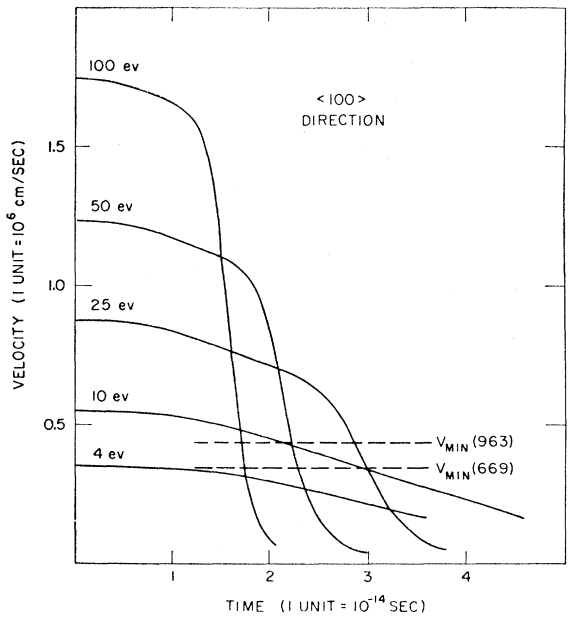

FIG. 4. Velocity of Cu atoms as a function of time for different initial velocities from the calculations of Vineyard *et al.*[8] The initial velocity is in the ⟨100⟩ direction. The horizontal lines marked $v_{\min}$ are the minimum velocity a recoil may have to contribute to the respective resonances.

in the crystal. Figure 5 presents slowing-down curves for 25 ev recoils in the three principal directions. Slowing down is most rapid at all energies in the ⟨110⟩ direction where the recoil aims at its nearest neighbor.

From the velocity-time data, the value of the resonance attenuation, $\alpha(\mathbf{v}_0)$, defined in Eq. (6) was calculated by graphical integration. The values of the state lifetimes used in these calculations are those presented in Table I and are based on the statistical factors derived assuming spin $\frac{1}{2}$ and $\frac{5}{2}$ for the 669-kev and 963-kev levels, respectively. Values of $\alpha(\mathbf{v}_0)$ and A obtained with lifetimes inferred using other spin values for these states will be discussed in Sec. VC. Since $\alpha(\mathbf{v}_0)$ is a function of the initial direction of the recoil in the lattice, results were obtained separately for the three principal directions and are presented as a function of initial velocity for both 669- and 963-kev resonances in Fig. 6. It should be noted that the lifetime of the

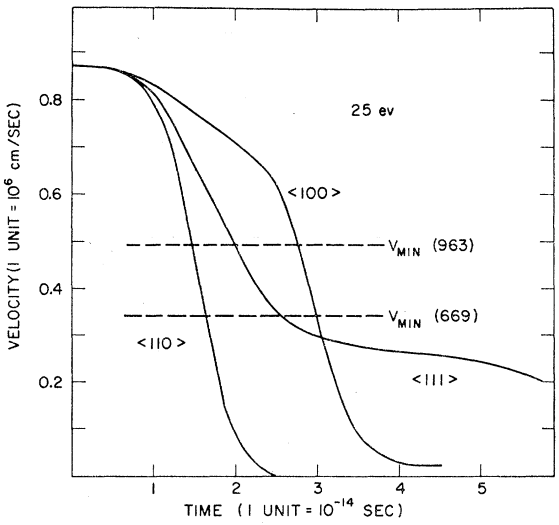

FIG. 5. Velocity as a function of time for Cu recoils with 25-ev initial energy and initial momenta along the indicated directions in the lattice.

state enters the calculation in two places. Since the time to drop below the resonance velocity is much smaller than the lifetimes, the exponential factor in Eq. (6) is not very different from unity, particularly for the 963-kev transition, and the main dependence of the attenuation is then nearly inversely proportional to the mean life of the state.

The weighting of $\alpha(\mathbf{v}_0)$ by the initial recoil spectrum as indicated in Eq. (7) was performed graphically. The function used for $P(v_0)$ was that given by Kofoed-Hansen.[21] This is an approximate form for the β-decay recoil spectrum. The approximation is that the Fermi function is constant (i.e., $Z=0$) which appears reasonable.[19] The results are presented in Table IV. To average over all direction of recoil, the statistical factors, 12, 6, and 8, for a face-centered cube were used for the

[21] A. Kofoed-Hansen, Phys. Rev. 74, 1785 (1948).

⟨110⟩, ⟨100⟩, and ⟨111⟩ directions. Analysis of the data for intermediate angles at 25 ev is not in disagreement with this method of averaging. Since the recoil velocities contributing to 963-kev resonance lie within the range of those contributing to the 669-kev resonance, errors in the averaging process tend to affect the calculations for both resonances in the same way.

C. Discussion of Solid Yields

The agreement of the values of A calculated from the model and those obtained from the experimental results is well within the errors indicated in Table IV. Since the error in the observed value reflects the error in the value of k while the error in the calculated value depends on the error in the mean life, the errors are not independent. If the experimental value of A is increased within the error, the calculated value must be correspondingly decreased. The calculated results in Table IV depend on the spin of the state since the actual lifetime of the state enters the calculation and the measurement of k gives only the product of a statistical factor and the lifetime. The observed value of A does not depend on the spin but only on the observed value of k.

TABLE IV. Resonance attenuations in solid copper.

	669 kev $\tau_m=(2.94\pm0.24)\times10^{-13}$	963 kev $\tau_m=(7.2\pm1.8)\times10^{-13}$
calculated		
$A\langle110\rangle$	0.074	0.021
$A\langle100\rangle$	0.106 ±11%	0.031 ±23%
$A\langle111\rangle$	0.109	0.030
A_{average}	0.092±0.010	0.026±0.006
Measured A	0.079±0.009	0.024±0.006

If we assume a spin $\frac{3}{2}$ for the 669-kev level, the value of τ_m becomes 5.9×10^{-13} sec and the calculated value of A is then 0.047 ± 0.005. This is clearly in disagreement with the measured value of A. Thus the yield measurements clearly favor spin $\frac{1}{2}$ for the 669-kev state in agreement with nuclear systematics as discussed in Sec. II.

The theoretical values of A for the 963-kev state for the spin assignments $\frac{1}{2}$ and $\frac{3}{2}$ are 0.078 ± 0.018 and 0.039 ± 0.009, respectively. It is seen that the agreement between the measured value and the theoretical value is most satisfactorily for the $\frac{5}{2}$ spin assignment in conformity with the unambiguous assignment from the Coulomb excitation angular correlation.

D. Resonance Yield from Aqueous Sources

Very little experimental evidence exists regarding the slowing down of 3–100 ev recoils in aqueous solution. As a first approximation we have considered the slowing down as the result of elastic collisions between the copper recoil and water molecules. If we assume that (1) all energy of the system is always in kinetic energy, (2) the Cu recoil has no angular momentum, (3) the water molecules are initially at rest, then such collisions can be characterized by a constant fractional velocity loss (when averaged over geometrical variables such as the impact parameter). Although this discrete collision model gives a step function for the velocity vs time, it is possible to smooth the function and characterize it by the differential equation

$$\frac{dv}{vds}=\frac{dv}{v^2dt}=-\frac{f}{D}\equiv-\frac{1}{L}, \tag{8}$$

where f is the average fractional velocity loss per collision and D is the mean distance the recoil moves between collisions. It is clear that $L=D/f$ is the characteristic path length required to reduce the recoil velocity to e^{-1} of its initial value.[22] Solution of the differential equation (8) gives the function $v(\mathbf{v}_0,t)=Lv_0/L+tv_0$ independent of the orientation of $\mathbf{v}_0$.

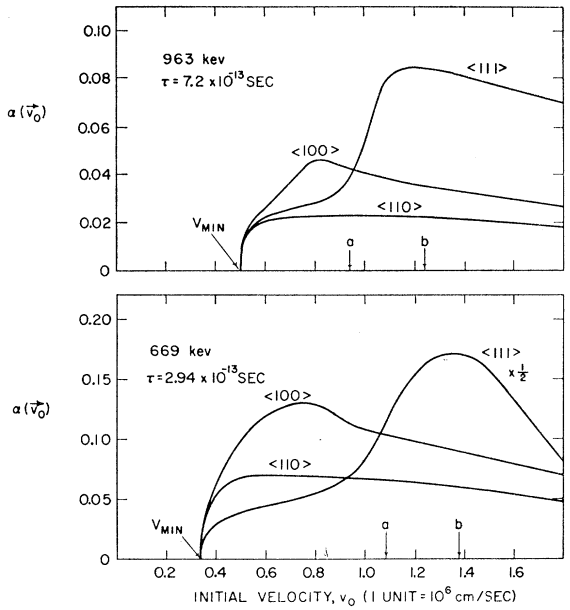

FIG. 6. The calculated differential attenuation of the resonance yield as a function of the initial recoil velocity for the solid Cu source. The two figures refer to the indicated resonances and τ is the mean life of the γ-emitting state used for the calculation. The arrows a and b refer to the maximum recoil velocity from the β decay and the K-capture recoil velocity, respectively.

[22] If the elastic collisions are considered to occur between spherical particles similar to billiard balls, the quantity f may be calculated. We assume that the water molecules are initially stationary and that the impact parameters are uniformly distributed on a plane normal to the recoil motion. In this case $f=|1-2(1-k^3)/3(1-k^2)|$ where $k=(M_1-M_2)/(M_1+M_2)$ and M_1 and M_2 are the masses of the recoil and environment molecules, respectively. For $M_1=63$ and $M_2=18$ we have $f=0.2$.

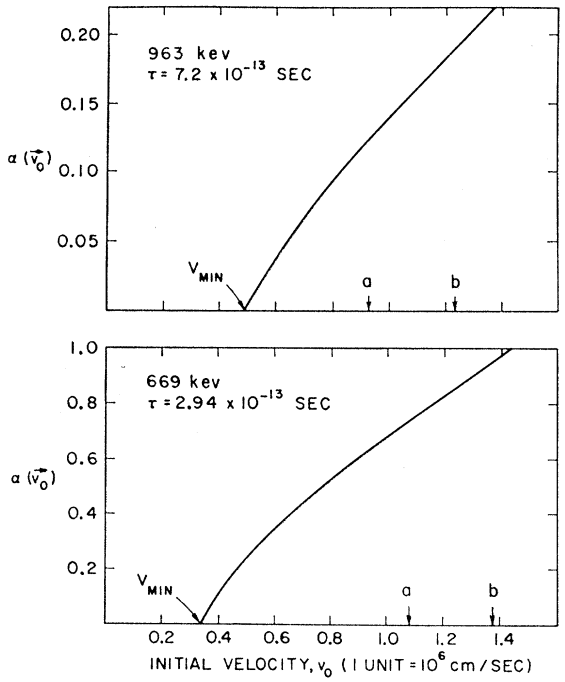

FIG. 7. The calculated differential attenuation as a function of initial recoil velocity for Cu in water. The quantity L has been taken as 6.8 A. The arrows a and b refer to the maximum recoil velocity from the β decay and the K-capture recoil velocity, respectively.

Substitution into Eq. (6) yields

$$\alpha(v_0)=\frac{v_0}{L}\left\{[\tau+L/v_0]-[\tau+L/v_{\min}]\exp\left[\frac{L}{\tau}\left(\frac{1}{v_0}-\frac{1}{v_{\min}}\right)\right]\right\}. \quad (9)$$

This function must be integrated numerically over the initial recoil spectrum as in Eq. (7) in order to find A.

The constant L was treated as an unknown and determined by fitting the calculated value of A to that measured for both Cu resonances. The values of L obtained in this manner were $(6.1_{-2.0}^{+2.5})$ A and $(7.9_{-3.0}^{+5.4})$ A for the 669- and 963-kev resonances, respectively. The errors on these values reflect the errors in the measured half-lives and resonance attenuations and include the correlation of these errors as discussed above. The ability to fit both attenuations with a common value of L such as 6.8 A lends credence to the use of Eq. (8). The function $\alpha(v_0)$ is plotted in Fig. 7 for the two copper resonances for this value of L. This is a reasonable value for the characteristic length in water. In particular, for the simple model of footnote 22, $D=(0.2L)=1.4$ A. The average spacing of H_2O molecules is $\sim$3.5 A thus yielding an average "collision radius" of 2.1 A for the sum of the copper and H_2O radii.

It is interesting to note that the contribution to the 669-kev resonance yield from gamma rays following the electron capture branch is almost identical from gas and liquid sources even though the recoil velocity in the liquid drops below $v_{\min}$ in approximately $\tau/2$. This is due to the increase in $N(E_r)$ as the recoil slows down in the liquid. If the recoil from the preceding radiation has a velocity several times $v_{\min}$ and the lifetime of the state is not too long, it is possible that the resonance yield from a condensed phase source will be greater than the yield from a gaseous source.[23]

To further test this model, we have also calculated the attenuation of the resonance yield for the 1.43-Mev resonance in Cr^{52}. The attenuation obtained from a water solution source was measured by Ofer and Schwarzschild[20] as $A=0.03\pm0.015$. The value of L was reduced from 6.8 A to 6.0 A for this calculation to account for the difference between the copper and chromium masses (according to the model of footnote 22). The result of this calculation yields $A=0.038\pm0.009$ in good agreement with measured value. This agreement is particularly significant since the effective recoils preceding the chromium resonance lie in the range 22 ev to 105 ev, considerably higher in energy than those in the copper cases.

VI. CONCLUSIONS

A. Transition Probabilities

The mean lives of the 669- and 963-kev states of Cu^{63}, the partial lifetimes for the $E2$ and $M1$ contributions to these transitions, and the ratios, F, of the transition probabilities to the single-particle estimates are presented in Table V. The partial $E2$ lifetimes have been deduced from the Coulomb excitation data.[3] The single-particle estimates have been obtained as indicated in the footnotes to the table; the spin factors have been included only for the $M1$ transitions.

The low-lying states of Cu^{63} have been characterized by Lawson and Uretsky[1] as arising from the coupling of the single $p_{\frac{3}{2}}$ proton with the semimagic Ni^{62} core. The ground state of Cu^{63} results from the coupling with the 0+ ground state of Ni^{62}. The first four excited states result from the various possible couplings of the odd particle with the 2+ state of the core. Some consequences of this model are: (1) The spin-weighted $(2j+1)$ center of gravity of the excited state energies in Cu^{63} should be equal to the energy of the 2+ state in Ni^{62}. (2) Gamma transitions from all of the four excited states to the ground state should be pure $E2$ and the reduced transition probabilities for each of these transitions should be equal to that of the $2+\rightarrow 0+$ transition in Ni^{62}.

It has been shown[1] that the center of gravity of the excited states of Cu^{63} is within 1% of the energy of the 2+ state of Ni^{62}. The reduced transition probabilities

[23] A similar enhancement has been observed by V. K. Rasmussen, F. R. Metzger, and C. P. Swann, Nuclear Phys. 13, 95 (1959). However, in this reference the recoils from a (p,p') reaction are approximately 100 times more energetic than those considered here.

TABLE V. Partial lifetimes.

E_r (kev)	$\tau_{measured}$ (sec)	$\tau(E2)$[a] (sec)	$F(E2)$[b]	$\tau(M1)$ (sec)	$F(M1)$[c]	$\delta^2=E2/M1$
669	$(2.94\pm0.24)\times10^{-13}$	3×10^{-11}	13	3×10^{-13}	0.20	0.01
963	$(7.2\pm1.8)\times10^{-13}$	5.1×10^{-12}	13	8.4×10^{-13}	0.04	0.16

[a] Calculated from $B(E2)$ of reference 3.
[b] Favored factor calculated relative to single-particle lifetime given by $\tau_{SP}=1.35A^{-4/3}E^5\times10^{-8}$ sec (E in Mev).
[c] Favored factor calculated relative to single-particle speed as given by S. A. Moszkowski (including proper statistical factor), in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955).

of the $E2$ part of both the 669- and 963-kev transitions are equal, within experimental errors ($\sim20\%$), to that observed in Ni^{62}.[24] It should be noted, however, that the $M1$ part of the 669-kev transition is retarded relative to the single-particle estimate only by a factor of five. Such a small hindrance of the $M1$ transition probability indicates a serious breakdown of the model.

B. Resonance Yield from Condensed Phases

In observation of nuclear resonance fluorescence utilizing the recoil momentum from a preceding radiation to obtain energy compensation, the yield is strongly dependent upon the source environment. The attenuation of the resonance yield from condensed sources is large for resonances involving states of lifetime greater than 10^{-14} sec. Utilizing a reasonable model for the trajectories of recoils in Cu metal, we have been able to calculate theoretical attenuation of the resonance yields which agree with those observed experimentally. The ratio of experimental to theoretical attenuation for the 669-kev resonance enables us to determine the spin of the 669-kev level as $\frac{1}{2}$.

Very little experimental information is known about the stopping of low-energy (3–100 ev) atoms in crystalline environments. The resonance fluorescence attenuation measurements are useful probes for determining the stopping properties of such recoils. Our attenuation measurements agree with the calculations based on the model of Vineyard *et al.*,[8] in which the force parameters were chosen by consideration of macroscopic properties of copper.

We have also performed calculations of the resonance attenuations for water solution sources in which it was assumed that the time rate of change of recoil velocity was proportional to the recoil energy. Using one adjustable parameter we have been able to fit the measured attenuations of both Cu resonances. A simple physical description of the slowing down process indicates that the parameter has a reasonable magnitude. This same model also gives good agreement with experiment for the 1.43-Mev resonance[20] in Cr^{52}.

The measurement of resonance attenuation can be used as a tool for investigating the slowing down in condensed media of "hot atoms" in an energy region almost inaccessible to other methods. In the present cases the necessity for averaging over a large range of initial velocities and directions obscures the details of the slowing down process. There are resonances, however, in which the recoil from the preceding radiation is monoenergetic. If the preceding radiation is a γ ray even the recoil direction may be defined. Investigation of such cases would be most interesting.

[24] L. K. McGowan and P. H. Stelson (private communication).

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