Gamma-Scintillations in Diphenylacetylene

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Koski and Thomas[1] have recently reported the observation of scintillations in diphenylacetylene and have estimated the signal-to-noise ratio of diphenylacetylene to be six times better than that of stilbene. Measurements at this laboratory also show diphenylacetylene to be a more efficient phosphor and indicate its luminescence decay constant at room temperature to be 8×10^{-9} sec., assuming a pulse whose amplitude decays as $e^{-t/\tau}$. This time measurement was made by a shorted coaxial line technique in which the output pulse generated by the photo-multiplier tube is partially canceled by its own out-of-phase reflection from the variable-length shorted line. The decay constant determined by this method has not been corrected for the inherent spread in transit time of the photo-multiplier tube.

The experiment was carried out with a 931-A photo-multiplier operating at 200 v per stage which was cooled by liquid nitrogen. A radium gamma-ray source was used for excitation.

The crystals were in flake form and were made by the Bios Laboratories, Inc., of New York City.

[1] W. C. Koski and C. O. Thomas, Phys. Rev. **76**, 308 (1949).

On the Isomerism of Kr^{83}

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In an investigation of D-bombarded Se, Langsdorf and Segrè[1] discovered the isomerism of Kr^{83} and estimated the energy of the conversion electrons to be about 35 kev. Measurements with the β-spectrometer by Helmholtz[2] indicated two soft γ-rays of energies 29 and 46 kev (Se+D sample).

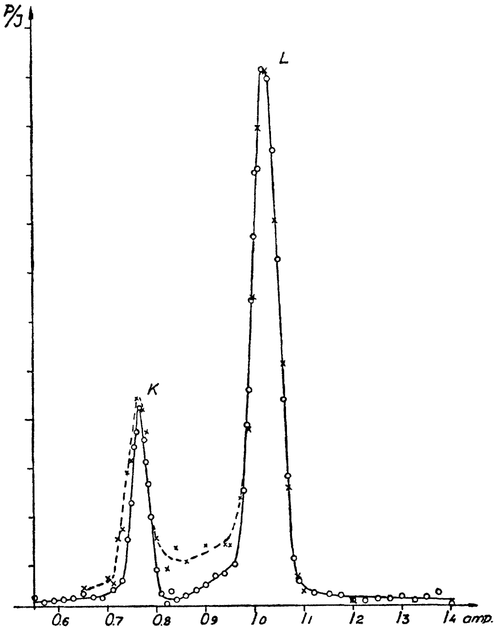

Fig. 1. Conversion lines of Kr^{83} (not corrected for absorption in the G-M tube window). Solid curve: Kr^{83} deposited on 0.15 mg Al/cm². Broken curve: Kr^{83m} deposited on 2.5 mg Al/cm².

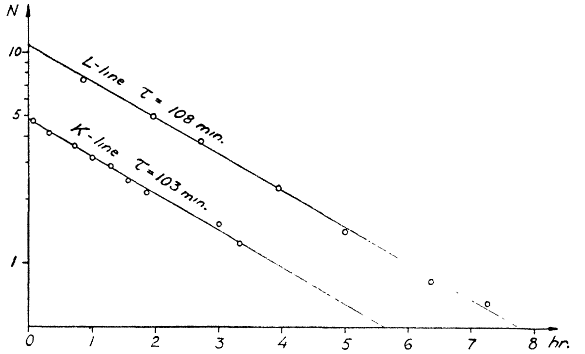

Fig. 2. Decay curves for the K- and L-lines measured in the β-spectrometer.

By separating the gaseous fission products of uranium (after cyclotron bombardment) in the electromagnetic isotope separator of the Nobel Institute,[3] it has been possible to obtain Kr^{83} with an activity sufficient for β-spectrometer investigations. This is an excellent way of preparing β-spectrometer samples. The active ions reach the collector foil with an energy of 30–60 kev and will in our case penetrate into the foil to a depth less than a hundred atomic layers[4] corresponding to an absorption thickness of the order of 1 μg/cm² in Al for the electrons from the deepest situated active atoms. It has proved possible to use 0.15 mg Al/cm² as collector foils. Thus, in most cases we obtain a mass-determined, carrier-free activity deposited on a very thin foil without any covering substance.

Table I. Conversion lines of Kr^{83}.

Conversion lines	$H\rho$ gauss-cm	$h\nu$ kev	Half-life (min.)	N_K/N_L line ratio	N_K/N_L surface ratio
K	446	31.5	103 ± 10	0.44 ± 0.06	0.39
L	598	32.5	108 ± 5		

The foil covered sample holder was fixed behind a circular slit ($r=5$ mm) in the collector arrangement of the separator and exposed to the "spot focused"[5] Kr^{83} line for about one hour. For mass number control the fission gases were mixed with stable Kr. A current of about 2 μamp. of Kr^{83} at 30 kev could be allowed during the separation without destroying the thin Al foil. The β-spectrometer data are summarized in Table I and Figs. 1 and 2.

Broken and solid lines refer to two different measurements with collector foils 2.5 and 0.15 mg Al/cm², respectively. The back-scattering effect of the thicker foil was too large to obtain an accurate value for N_K/N_L.

The absorption effect of the G-M tube window was calculated from measurements of the Co^{60} spectrum. The correction factor for N_K/N_L was 1.55 giving $N_K/N_L=0.44$, when comparing the ratio of the line heights and 0.39 for the line surfaces. The measurements were repeated in another lens spectrometer with a somewhat thinner G-M foil and $N_K/N_L=0.40$ was obtained for the ratio of the line heights. The $H\rho$-values in this case were 453 and 603 corresponding to $h\nu=32.0$ and 32.9 kev. Four measurements in two β-spectrometers gave the values 32.5, 32.5, 32.7, and 32.9 kev (mean value 32.7±0.5 kev) for the γ-energy from the L-line, which is much more accurately determined than the K-line.

Our γ-energy value of 32.7 kev probably corresponds to that of 29 kev reported by Helmholtz. It was not possible, however, to find the other converted γ-ray of 46 kev and, if it at all belongs to Kr^{83m}, the intensity of the conversion lines must be less than 1/20 of the L-line found by us. It is therefore possible that the 46-kev line belongs to Br^{83}.

The formula for the half-life given by Bethe and corrected for the theoretically calculated conversion coefficients (Hebb and Nelson) gives the half-life 2×10^{-1} sec. for $l=3$ and 3×10^5 sec. for $l=4$ (experimental value 6.5×10^3 sec.). According to the curves of Flügge,[6] $N_K/N_L\approx0.1$ for $l=4$ and $N_K/N_L\approx0.4$ for $l=3$ (electric radiation). The recently published curves of Tralli and Lowen[7] give $N_K/N_L\approx3$ for $l=3$ and magnetic radiation. For electric octupole radiation the agreement between the experimental value 0.44 of N_K/N_L and the theoretical one is very good. The half-life and internal conversion coefficients, however, indicate a mixture of ≈70 percent electric 2^4-pole and ≈30 percent magnetic 2^3-pole radiation. This would then still be an example of a parity forbidden isomeric transition.[8]

[1] A. Langsdorf, Jr. and E. Segrè, Phys. Rev. **57**, 105 (1940).
[2] A. C. Helmholz, Phys. Rev. **60**, 415 (1941).
[3] Bergström, Thulin, Svartholm, and Siegbahn, Arkiv f. Fys. **1**:11, 281 (1949).
[4] T. Koch, Nature **161**, 566 (1948).
[5] I. Bergström and S. Thulin, Phys. Rev. **76**, 1718 (1949).
[6] S. Flügge, Physik. Zeits. **42**, 227 (1941).
[7] N. Tralli and I. S. Lowen, Phys. Rev. **76**, 1541 (1949).
[8] P. Axel and S. M. Dancoff, Phys. Rev. **76**, 892 (1949).

The Effect of Electronic Paramagnetism on Nuclear Magnetic Resonance Frequencies in Metals

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The nuclear paramagnetic resonance frequency for an atom in the metallic state usually is appreciably greater than the resonance frequency when the atom is in a non-metallic compound.[1] These frequency shifts are of the order of a few tenths of one percent, and are hence much too large to be accounted for by a simple difference in magnetic susceptibility of the materials or by differences in diamagnetic correction for the metallic and non-metallic atoms. It is proposed that such shifts are primarily due to orientation by the magnetic field of the spins of conduction electrons near the top of the Fermi distribution, and the interaction of these electrons with the nuclei. Since the conduction electrons usually have a very large probability density near the nucleus, this may be understood as an enormous concentration of the local magnetic susceptibility of the metal in the vicinity of the nuclei. It may also be compared with and calculated from the magnetic hyperfine structure interactions in an isolated atom. Diamagnetic effects of the same type appear to be very much smaller since the electronic currents responsible for the diamagnetism of the conduction electrons are appreciable only in the outer regions of the atom.

The fractional shift in nuclear resonance frequency between metal and non-metal is simply the fractional amount by which the presence of the conduction electrons increases the mean magnetic field strength H at the nucleus. Neglecting the small diamagnetic effect, we consider only the field ΔH due to the electron spins, which is $8\pi/3$ times the mean density of spin moment at the nucleus, assuming cubic symmetry. If the electron distribution is not cubically symmetric about the nucleus, then electron density at points other than at the nucleus contributes to ΔH, and ΔH will depend to some extent on orientation of the crystal with respect to H. In terms of wave functions of individual electrons in the metal the spin moment density at the nucleus can be written $\chi_pMH\langle|\psi_F(0)|^2\rangle_{\text{Av}}$, where χ_p is the spin contribution to the macroscopic susceptibility per unit mass, M is the mass of one atom, and $\langle|\psi_F(0)|^2\rangle_{\text{Av}}$ is the average probability density at the nucleus for all electronic states on the Fermi surface. Now the hyperfine structure splitting $\Delta\nu$ for an s electron in the free atom is proportional to $|\psi_a(0)|^2$, where ψ_a is the wave function of the s electron in the free atom. Dividing the above expression for ΔH by the familiar Fermi expression for $\Delta\nu$ gives the fractional frequency shift in terms of a ratio of metallic and atomic wave functions:

$$\frac{\Delta H}{H}=\frac{hc\Delta\nu I\chi_pM}{\mu_I\mu_e(2I+1)}\frac{\langle|\psi_F(0)|^2\rangle_{\text{Av}}}{|\psi_a(0)|^2} \tag{1}$$

where $hc\Delta\nu$ is the hyperfine splitting in energy units, μ_e is the Bohr magnetron, μ_I is the nuclear moment, and I the nuclear spin.

TABLE I. Observed shifts of nuclear resonances in metals due to free electron paramagnetism, and comparison with theory.

Nucleus	H.f.s. splitting $\Delta\nu$, in cm^{-1} for s electron	Observed shift $\Delta H/H$, (percent)	χ_p per gram $\times10^6$	$\chi_p\langle\lvert\psi_F(0)\rvert^2\rangle_{\text{Av}}/\lvert\psi_a(0)\rvert^2\times10^6$	
				theory	from (1)
Li^7	0.027	0.04	3.0[a]	~2.3	2.6
Na^{23}	0.060	0.10	0.98[b]	0.80	0.63
Cu^{63}	0.39	0.23	~0.11[c]		0.078
Be^9	0.040	<0.002	~0.30[d]	~0.15	<0.025[e]
Pb^{207}	2.1[f]	1.2	~0.10[c]		0.0091
Al^{27}	0.35[f]	0.16	~0.47[c]		0.21
Ca^{69}	0.58[f]	0.44	~0.20[c]		0.083

[a] Measured value with diamagnetic corrections, S. R. Rao and K. Sarithri, Proc. Ind. Acad. **6**, 207 (1942).
[b] Average of measured values with diamagnetic corrections, F. Bitter, Phys. Rev. **36**, 978 (1930).
[c] Calculated value assuming free valence electrons.
[d] Theoretical value (reference 3).
[e] The small diamagnetic contribution to the shift may increase this limit by a factor of 2 or so.
[f] Estimated from Goudsmit's formula and measured values for ionized atoms.

We have attempted to calculate the theoretical value of the last factor in (1) for the cases (Na, Li, Be) for which adequate calculations of metallic wave functions are at present available. This factor may be written $(|\psi_0(0)|^2/|\psi_a(0)|^2)\bar{s}_K$, where ψ_0 is the wave function of an electron at the bottom of the conduction band and $\bar{s}_K$ is the mean over the Fermi surface of

$$s_k=(|\psi_k(0)|^2)/(|\psi_0(0)|^2).$$

Calculations by the Wigner-Seitz method[2] give ψ_0; $\bar{s}_k$ can be determined in either of two ways. The first, suitable only for monovalent metals, is to assume that s_k is adequately represented by the first two terms of its development in powers of the square of the electronic wave vector k:

$$s_k=1+k^2ds_k/d(k^2)+\cdots, \tag{2}$$

and to use the expression given by Herring and Hill[3] for $ds_k/d(k^2)$ (Appendix III of this reference; a factor 6 should be inserted before α in Eq. (M).) This method was applied to the case of Na with the aid of wave functions kindly supplied by Dr. Bardeen, and gave $\bar{s}_k=0.70$. Similar calculations were made for Li, but no results will be quoted because it was found that the available wave functions were based on an incorrect potential field for the Li ion. Comparison with the case of Be, to be discussed next, suggests that $\bar{s}_k$ for Li may be of the order of 0.4. For Be, which is divalent, the first two terms of (2) do not give at all a good approximation, and a second procedure was therefore adopted. This consisted in dividing up the Fermi surface into six portions, estimating an average s_k for each portion from the values of s_k directly calculated for states near the Fermi surface (Table VII of reference 3), and combining with suitable weights. This gave $\bar{s}_k=0.32$, and this value did not seem to be particularly sensitive to changes in the technique of averaging.

The quantity $|\psi_0(0)|^2/|\psi_a(0)|^2$ has approximately the values 1.16, 1.9, and 1.6, for Na, Li, and Be, respectively. For other metals its behavior can be inferred qualitatively by noting how much of the charge of the outermost s electrons of the free atom lies outside a sphere whose volume equals the atomic volume of the metal; it appears from this that $|\psi_0(0)|^2/|\psi_a(0)|^2$ should be appreciably smaller for such multivalent metals as Al, Ga, and Pb than for the mono- and divalent ones.