k the k'th electron with spin $\mathbf{S}_k$, V is the total electrostatic interaction, and $\mathbf{r}_{kl}$ is the vector $\mathbf{r}_k-\mathbf{r}_l$. $\mathfrak{H}_{II}$ corresponds to the direct magnetic interaction of the two nuclei and reduces to zero when averaged over all molecular rotational states. The remaining quantities are defined in the references.[2-4]

With the aforementioned Hamiltonian, the energy of the system can be calculated to second order by perturbation theory. The terms which depend on the products of the spins of the two nuclei then correspond to the various spin-spin interactions. The terms arising from the bracketed quantity in $\mathfrak{H}_1$ are analogous to the magnetic shielding terms calculated by Ramsey[2] for uniform magnetic fields and are the ones considered by Hahn and Maxwell[1] and found to produce too small an effect. The terms suggested in the present paper are the second-order perturbation terms arising from $\mathfrak{H}_2$ and $\mathfrak{H}_3$ and their cross products. The effects of these terms have been calculated approximately for molecular HD with the use of Heitler-London wave functions. The frequency shift arising from second-order perturbations by $\mathfrak{H}_2$ was found to be 0.5 $\mathbf{I}_H\cdot\mathbf{I}_D$ cycles per second when averaged over all molecular rotational states, which is small and of the same order of magnitude as the terms considered by Hahn.[1] The cross terms between $\mathfrak{H}_2$ and $\mathfrak{H}_3$ vanish when averaged over all molecular rotational states. However, the second-order perturbations by $\mathfrak{H}_3$ give a frequency shift that is relatively large and approximately equal to 70 $\mathbf{I}_H\cdot\mathbf{I}_D$ cycles per second. It is of interest to note that this term, which is the important one, can be written approximately as $0.46\ \mathbf{I}_H\cdot\mathbf{I}_D\Delta\nu_H\Delta\nu_D/(\Delta E/\hbar)$ where the $\Delta\nu$'s are the atomic hyperfine separations, and ΔE is the separation of the lowest singlet and lowest triplet states in molecular hydrogen.

Although a similar calculation in the case of dichloroacetaldehyde is precluded by the lack of a suitable wave function for the molecule, it is quite reasonable from the foregoing calculation that an effect as large as the observed 0.7 cycle per second should be observed since the exchange coupling of the electrons with each other in the successive bonds lying between the two hydrogens is large. In some substances much larger interactions than the aforementioned could probably be obtained if ΔE were suffiently small.

Experiments to observe this effect in a simple molecule like HD are being planned by the authors. Although such an interaction should also occur in homonuclear molecules such as H_2, it would not be observable in transitions involving no change in molecular rotational state. The authors wish to express their appreciation to Dr. Hahn for sending them an advance copy of the manuscript of his very interesting paper.[5]

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[5] Subsequent to the submission of this letter the authors have studied the recent article of H. S. Gutowsky and C. J. Hoffman [J. Chem. Phys. **19**, 1259 (1951)] in which these authors find multiplet resonance lines with PF_3. The mechanism suggested in the present letter should account for a multiplet structure of the observed nature. This mechanism should produce a larger effect than the one considered by Gutowsky, McCall, and Slichter [Phys. Rev. **84**, 590 (1951)] which depends only on electronic orbital contributions arising from $\mathfrak{H}_1$ in Eq. (2) above.

Possible Natural Radioactivity of Neodymium*

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A NUMBER of investigators have looked for natural radioactivity in neodymium.[1-12] All agree that alpha-activity is absent, and most either reported no beta-activity or ascribed the observed activity to common radioactive impurities. However, Libby,[7] who used by far the most sensitive detection method, reported a beta-radiation of maximum energy of about 11 kev and specific activity about 60–120 disintegrations per second per gram. That this activity is truly characteristic of neodymium has been in doubt because Libby did not repurify or fractionate his samples from all other elements to constant specific activity. Takvorian[9] reported inability to detect activity in neodymium, but an inspection of his data shows that his sensitivity was grossly inadequate. A reference by Broda[11] to the unpublished work of Jha (1949) gives no data but implies confirmation of Libby's findings. A recent report by Curran, Dixon, and Wilson[12] states that Libby's data are probably wrong and that the maximum beta-energy may greatly exceed 11 kev. The implication is that radiation attributable to the element was observed, but no indication of the level of activity or degree of chemical purification is given.

One of us[13] has pointed out that Nd^{150} appears to be outside the limits of beta-stability for even-even nuclides, and suggested the possible identification of the radiations found by Libby with those of Nd^{150}. This would imply the natural occurrence of Pm^{150} in equilibrium with Nd^{150} in rare earth minerals, a relatively long lifetime for the daughter being required to explain the absence of hard radiations in Libby's samples. An alternate possibility suggested, that Nd^{140} is long-lived and occurs naturally, has been eliminated by the subsequent finding[14] that Nd^{140} has a 3.3-day half-life with a 2-minute daughter emitting energetic radiations. The determination[15] that Pm^{145} undergoes electron-capture decay to Nd^{145} removes the possibility[16] that the latter is unstable.

Our measurements were made with a proportional counter accommodating internally solid samples 1650 cm² in area, filled with 68-cm argon and 8-cm ethylene. About 10 percent of the sample is obscured by three longitudinal rods which support the wire from one end so that samples can be inserted from the other end. It is used as a crude spectrometer for low energy radiations by varying the external amplifier gain so as to obtain bias curves. An energy calibration was obtained from the sharp cutoff of the manganese K x-rays of Fe^{55} (6 kev), and the highest gain used corresponds to a bias level of 0.3 kev on the energy scale. By combined absorption and bias-curve measurements of the K Auger electrons from this transition (5 kev), we have verified that electrons of a very few kev energy are counted with apparent high efficiency and no reduction in energy. The background has been reduced to about 32 counts per minute by shielding with a ring of Geiger counters in anticoincidence, 2 inches of hot-rolled steel, and 3 inches of lead. Since Libby observed 10 counts per minute from an area of 193 cm² at 14 percent geometry, we would expect 270 counts/min from our 1650 cm² at 45 percent geometry if his data were correct, the sample thickness being immaterial for such low energy radiations.

Commercial 99 percent pure neodymium oxide (Lindsay Light and Chemical Company, Chicago) was twice purified in a cation exchange column (Dowex 50) 80 cm long, with very slow citrate elution, scavenged by barium sulfate and zirconium iodate precipitations, precipitated as oxalate, and ignited. A 15-gram layer of the resulting oxide spread uniformly on a copper sheet gave a counting rate about 4 per minute above background at the lowest bias. The bias curves showed that practically all of the excess radiations were more energetic than 11 kev. Moreover, the sensitivity of the copper-lined counter to external gamma-radiation (Co^{60}) is increased about 11 percent by the Nd_2O_3 coating, which just about accounts for the excess observed, since most of the residual background is presumably due to gamma-radiation. No growth of activity was observed in two months. We conclude that not more than 1 count/min of either soft or hard radiation is contributed by the neodymium. Accordingly, the maximum specific beta-activity is 0.003 disintegration per second per gram of element, corresponding to a minimum half-life of 2×10^{15} years for Nd^{150}.

In spite of the negative results of this experiment we feel that it is still likely that Nd^{150} is unstable but with a highly forbidden decay and very long lifetime. In the discussion of the limits of beta-stability,[13] Nd^{150} and Ca^{48} were given similar special mention as being apparently outside of the stability region, and another

letter[17] cites evidence that Ca^{48} is indeed unstable but with an extremely long lifetime. Nordheim[18] has pointed out that odd-odd nuclides frequently have large nuclear spins, and this may well be the case for Pm^{150}. Nd^{150} could probably decay only to the ground state of Pm^{150}, and the large spin change would make this transition highly forbidden as must be the case for Ca^{48}.

Although we did not look specifically for alpha-activity, our measurements set an upper limit of 0.02 alphas per second per gram of neodymium, about one-fifteenth of the previous limit set by Bestenreiner and Broda.[10]

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The Hyperfine Structure Anomaly of the Rb Isotopes*

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Effects arising from the distribution of the nuclear magnetic moment over a finite volume lead to a discrepancy in the ratios of the hfs splittings of Rb^{85} and Rb^{87}, as measured directly[1] and as calculated from the ratio of the nuclear moments, if the nuclei are assumed to be point dipoles.[2] As the effect depends strongly on the relative distribution of the spin and orbital magnetic moments within the nuclear volume of each isotope, it presents a critical test of a nuclear model and accurate knowledge of its magnitude affords a valuable guide to theoretical investigation. The magnitude of this anomaly has been explained with some success by considering the nuclei in question to consist of a single nucleon moving in the field of a nuclear core of no intrinsic angular momentum.[3,4]

Bitter, using his own results[2] and data obtained by Millman and Kusch,[1] showed that

$$\Delta \equiv \frac{(\Delta\nu^{87}/\Delta\nu^{85})_{\text{obs}} - (\Delta\nu^{87}/\Delta\nu^{85})_{\text{calc}}}{(\Delta\nu^{87}/\Delta\nu^{85})_{\text{calc}}},$$

is (0.33±0.05) percent. The recent direct measurement by Yasaitis and Smaller[5] of the ratio $\mu(Rb^{87})/\mu(Rb^{85})$ is of much greater accuracy (one part in 250,000) than either the previous moment ratio or the previous ratio of the hfs splittings. It became worthwhile, therefore, to redetermine the $\Delta\nu$ ratio to a precision comparable to that of the moment ratio and to obtain Δ to a high degree of precision.

We have used a previously described[6] atomic beam method to determine $\Delta\nu$ from a measurement of some of the lines $\Delta m_I = \pm 1$, $\Delta m_J = 0$ in the hfs spectrum of the Rb isotopes. Those transitions were chosen which give rise to doublets ($m_J = \pm 1/2$) whose mean frequencies attain maximum values at characteristic values of x, where $x \equiv (g_J - g_I)\mu_0 H/h\Delta\nu$. $Rb^{87}(I=3/2)$ has one such doublet, while $Rb^{85}(I=5/2)$ has two. Within a small range of x, bracketing the value for maximum frequency, the line frequencies are almost field independent. The measured doublet separation ($2g_I\mu_0 H/h$) and approximate values of g_J/g_I and $\Delta\nu$ then suffice for the determination of x, which, together with the measured mean frequency, yields a value of $\Delta\nu$ whose accuracy is limited only by the precision of the frequency measurements. Of the two maxima in the Rb^{85} spectrum one (m_F: $0 \leftrightarrow 1$) occurs at an x of 5.8284 and the other (m_F: $-1 \leftrightarrow -2$) at an x of 1.9250. The first of these, therefore, corresponds to a larger doublet separation and yields a more precise value of x. In addition for the doublet (m_F: $0 \leftrightarrow -1$), the frequency at values of x close to that at the maximum mean frequency is much less field dependent than the frequency of the doublet (m_J: $-1 \leftrightarrow -2$) near its maximum mean frequency. Thus, the doublet (m_F: $0 \leftrightarrow -1$) is much better suited for an accurate determination of $\Delta\nu(Rb^{85})$ and was used in this experiment.

Our results are

$$\Delta\nu(Rb^{85}) = 3035.730 \pm 0.005 \text{ Mc/sec}$$

$$\Delta\nu(Rb^{87}) = 6834.681 \pm 0.010 \text{ Mc/sec}.$$

Measurements taken for the transition (m_F: $-1 \leftrightarrow -2$) gave

$$\Delta\nu(Rb^{85}) = 3035.745 \pm 0.025 \text{ Mc/sec},$$

which compares favorably with the forestated value. The ratio

$$\Delta\nu^{87}/\Delta\nu^{85} = 2.251413 \pm 0.000005,$$

together with the result of Yasaitis and Smaller,[5] yields

$$\Delta = (0.3501 \pm 0.0006) \text{ percent}.$$

Bohr and Weisskopf[3] found the best agreement between the experimentally determined value of Δ and that predicted from their rather crude nuclear model by choosing for g_S and g_L the g-values of the odd proton. They thus calculated $\Delta = 0.29$ percent. Bohr subsequently studied a model in which he assumed the individual nucleons to move in an asymmetric average field[7] and used it to recalculate the hfs anomaly in greater detail.[4] Neglecting the contribution of the nuclear core to the angular momentum and making special assumptions about the couplings between the various angular momentum vectors, he found $\Delta = 0.26$ percent. The increase in the experimental value of Δ here reported as compared to the earlier results, and the decrease in the value calculated by Bohr on the basis of a refined nuclear model, lead to a rather large discrepancy between Δ_{obs} and Δ_{calc}. It is clear that a more detailed theoretical analysis is required to predict a value of Δ in good agreement with and comparable in uncertainty to Δ_{obs}.

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Condition for Radiation from a Solar Plasma

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Recent attempts to account for the bursts of radiofrequency electromagnetic radiation received from the sun have employed the mechanism of wave growth in a uniform plasma of infinite extent for the conversion of the kinetic energy of moving streams into the time varying fields of a space charge wave. The existence of a Poynting flux vector within the infinite oscillating plasma has then been taken as the criterion for the escape of radiation.[1] It is the purpose of this note to point out that while a model which is infinite in extent is undoubtedly satisfactory for the determination of the bands within which wave amplification can occur in the interior of a finite sized plasma, one cannot justify the use of a point value of the Poynting vector within a uniform infinite plasma as a criterion for the reception of radiation at a point exterior to, and far removed from, the actual oscillating plasma.