of the components in the oven are Pyrex—there does not appear to be any physical reason why the technique could not be extended to high temperatures.

Optical pumping utilizing the alkalis has in the past provided very precise values for free-atom hfs and g_J values. The high precision is especially useful in looking for effects caused by higher order multipole interactions. The present technique will make possible the pumping of a considerable number of new elements—As, Sb, Bi, Cr, Mn, Mo, Tc, Re, Cu, Ag, Au, Eu, and Am all have orbital S states with $J \neq 0$ and therefore can potentially be polarized by spin-exchange collisions.

Several aspects of interatomic interactions have been studied with optical pumping. These include pressure shifts of hfs[10] and g_J values,[11] spin-exchange cross sections,[2-5] spin-relaxation cross sections, and diffusion coefficients.[12] The ability to measure these quantities over an extended temperature range should greatly assist in their understanding. Of particular interest is the energy dependence of the electron-alkali spin-exchange cross section.[13] It is also possible that the utility of optical pumping in chemistry may be increased.

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DETERMINATION OF MUONIUM HYPERFINE STRUCTURE INTERVAL THROUGH MEASUREMENTS AT LOW MAGNETIC FIELDS*

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The ground-state hyperfine structure interval $\Delta\nu$ of muonium has been remeasured with significantly improved accuracy through the observation of transitions between hfs states at low magnetic fields. The formation of muonium has been observed for the first time in krypton and the fractional pressure shift of $\Delta\nu$ has been measured in this gas. The average value of $\Delta\nu$ obtained from these measurements is $\Delta\nu = 4463.26 \pm 0.04$ Mc/sec.

The hyperfine structure interval $\Delta\nu$ of the muonium atom (μ^+e^-) in its ground state has been remeasured with improved accuracy through the observation of transitions between magnetic sublevels at weak magnetic field (about 3 G)[1] and at very weak magnetic field (about 10 mG). Compared with the earlier high-field experiment,[2,3] these measurements involve some different experimental techniques and thus provide an independent determination of $\Delta\nu$. The more accurate value for $\Delta\nu$ can be used to obtain a value for the fine structure constant α, or to determine a value for the muon magnetic moment, μ_μ, free from uncertainties of magnetic shielding.[4]

In a low external static magnetic field H, the relative energies of the sublevels of muonium are

given with adequate accuracy in frequency units by the equations[3]

$$\nu_{F=0,M_F=0} = 0, \quad \nu_{1,1} = \Delta\nu + z,$$

$$\nu_{1,0} = \Delta\nu, \quad \nu_{1,-1} = \Delta\nu - z,$$

where the Zeeman term is $z = (g_J\mu_0{}^e + g_\mu\mu_0{}^\mu)H/2h$ in which g_J = electron g value and g_μ = muon g value. Under weak-field conditions ($H \simeq 3$ G) the transitions $(F, m_F) = (1,1) \leftrightarrow (0,0)$ and $(1,-1) \leftrightarrow (0,0)$ are experimentally well resolved, since $z \simeq 4$ Mc/sec. At very weak field ($H \simeq 10$ mG) these transitions are unresolved since $z \simeq 0.01$ Mc/sec and the observed resonance linewidth is greater than 0.5 Mc/sec. The smallness of the Zeeman term at low magnetic field provides a distinct experimental advantage over measurements at a high magnetic field.

The theory of the resonance line shape for the weak-field case involves only two levels and has been given.[2,3] In the very weak field case, the line-shape theory must consider the three levels $(F, m_F) = (1,1)$, $(1,-1)$, and $(0,0)$. Solutions have been obtained for the state amplitudes in this case. The resulting line shapes are Lorentzian with increased widths and heights as compared with the two-level case. The level $(F, M_F) = (1,0)$ does not enter the calculation under the assumption that the time-varying magnetic field is perpendicular to the static field.

The experimental method is similar in principle to that of the high-field experiment. Polarized muonium is formed by stopping muons from the Nevis synchrocyclotron in a high-pressure noble-gas target (See Fig. 1). The microwave-induced changes in the populations of the magnetic levels are observed through the resulting change in the angular distribution of the decay positrons. A structure consisting of three large Moly-Permalloy shields surrounding a solenoid and associated correction coils was used to generate the necessary static fields of 2.85 and 0.010 G. For the very weak-field case the solenoid and coils were disconnected, and the outer shield was equipped with Mumetal end caps. The maximum instability and inhomogeneity of the field over the target region were ±1 and ±3 mG, respectively. At weak field the static magnetic field was mapped and monitored with a Rb^{85} optical-pumping magnetometer. At very weak field a fluxgate magnetometer was used. The microwave frequency was harmonically generated from a crystal oscillator with a frequency stability of better than 1 part in 10^7. After amplification this signal was used to drive a high-Q cavity in the TM_{220} mode in which the time-dependent magnetic field was perpendicular to the static field H. A fast feedback system

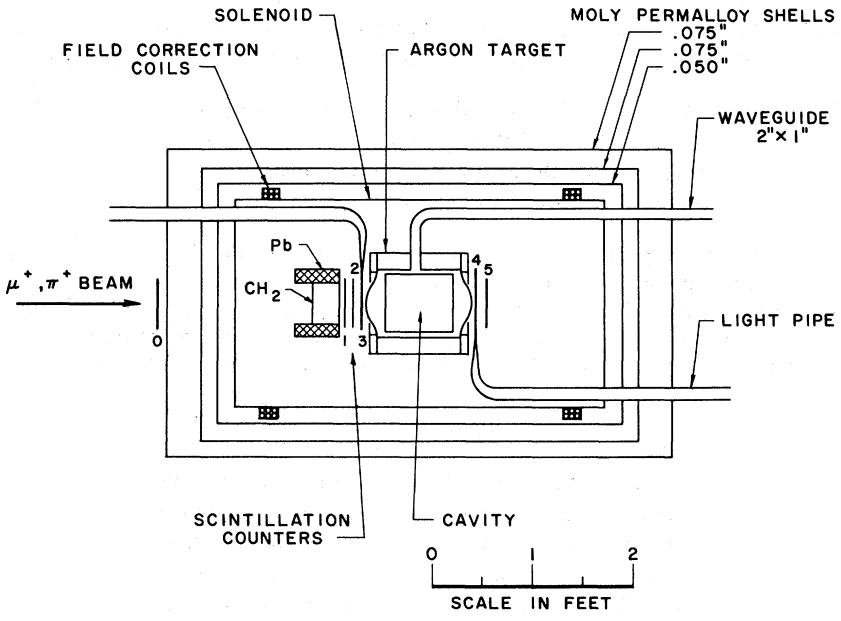

FIG. 1. Schematic diagram of experimental arrangement for observing muonium hfs transitions at low magnetic fields.

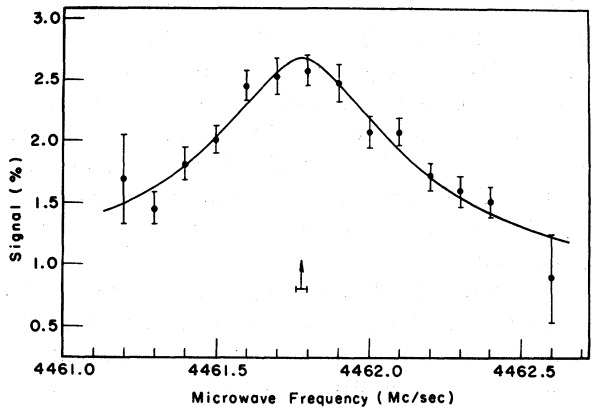

FIG. 2. Resonance curve observed at $H = 0.010$ G for the unresolved transitions $(F, M_F) = (1, -1) \leftrightarrow (0, 0)$ and $(1, 1) \leftrightarrow (0, 0)$ in krypton buffer gas at 32 800 Torr. The error bars (1 standard deviation) are due to counting statistics. The solid curve is a best fit to a theoretical Lorentzian line shape. The fitted center is 4461.779 ± 0.017 Mc/sec.

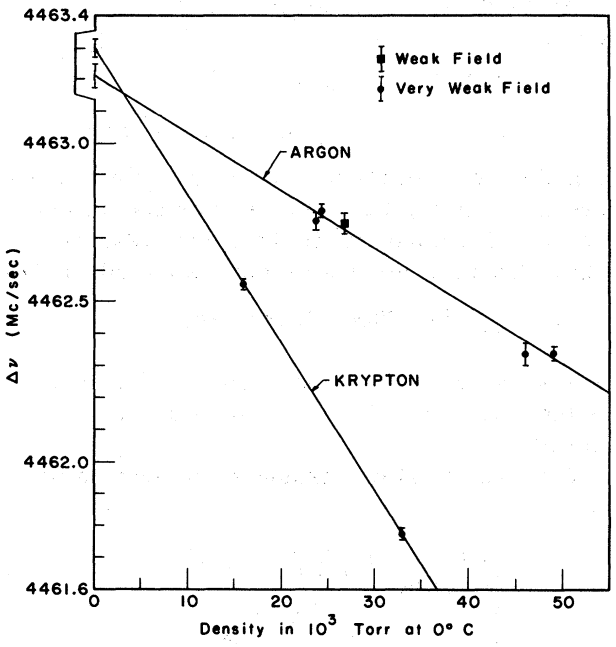

FIG. 3. Observed values of $\Delta\nu$ versus gas density for Ar and Kr. The solid curves are straight lines fitted to the data. The density has been corrected for the departure from ideal-gas behavior. The points at zero density are the extrapolated intercepts and not measured values.

was used to stabilize the microwave power level to 2%. The resonance line was observed by varying the microwave frequency. The temperature and pressure of the buffer gas were monitored continuously with uncertainties of ±0.5°C and ±1 psi, respectively.

Data were taken with both argon and krypton at two different pressures. Figure 2 shows an observed resonance curve at very weak magnetic field obtained in about 100 h of beam time. The experimental data fit Lorentzian line shapes very well. Increasing the gas density increases both the stopping rate and the uncertainty introduced by the pressure-shift extrapolation; for these reasons pressures in the range 20-60 atm were used. Figure 3 shows a plot of all the weak- and very weak-field data.

A value for the fractional pressure shift (FPS) $[\mathrm{FPS} = (1/\Delta\nu)\partial\Delta\nu/\partial P]$ may be obtained from the slope of the straight-line fit to the observed points. The values may be compared with the FPS values for hydrogen in the same buffer gases. For muonium[5] in argon at high field the FPS $= -4.05 \pm 0.49$; at low field the FPS $= -4.07 \pm 0.25$. For hydrogen in argon[6] the FPS $= -4.78 \pm 0.03$. For muonium in krypton the FPS $= -10.4 \pm 0.30$. For hydrogen in krypton[7] the FPS $= -10.4 \pm 0.20$. All FPS values are in units of 10^{-9}/(Torr at 0°C). The apparent difference between the FPS values for muonium and hydrogen in Ar may be evidence of an isotope dependence of the FPS[8] or of a nonlinearity in the density dependence of $\Delta\nu$. There is excellent agreement between the FPS values for muonium and hydrogen in Kr.

A value of $\Delta\nu$ for free muonium is obtained from the intercept of the straight-line fit to the data. The results are

$$\Delta\nu = 4463.302 \pm 0.027 \text{ Mc/sec (from Kr data)},$$

$$\Delta\nu = 4463.220 \pm 0.033 \text{ Mc/sec (from Ar data)}$$

in which the errors (1 standard deviation) are statistical counting errors. The sources of systematic errors considered and estimates (at least 1 standard deviation) of their effect on the determination of $\Delta\nu$ are as follows: (1) uncertainty in microwave power level in cavity, 5 kc/sec; (2) uncertainty in microwave frequency, 0.1 kc/sec; (3) instability and inhomogeneity of static magnetic field, 3 kc/sec; (4) uncertainties in buffer-gas pressure and temperature, 3 kc/sec; and (5) approximations in derivation of theoretical line shape, 3 kc/sec. These errors are negligible compared with the counting errors.

The value of $\Delta\nu$ from the previous high-field measurement is[2]

$$\Delta\nu = 4463.15 \pm 0.06 \text{ Mc/sec}.$$

A weighted average of the three measurements

(high-field Ar, low-field Ar, and low-field Kr) gives

$$\Delta\nu = 4463.255 \pm 0.020 \text{ Mc/sec}$$

(1 standard deviation).

The difference of about 2 standard deviations between the Ar and Kr measurements may be evidence of a nonlinearity in the argon pressure shift. To allow for this we take as a final number

$$\Delta\nu_{\text{expt}} = 4463.26 \pm 0.04 \text{ Mc/sec},$$

where the error (1 standard deviation) assigned is the variance of the three values about their mean.

The theoretical expression for $\Delta\nu$ may be written[3]

$$\Delta\nu = \left(\frac{16}{3}\alpha^2 cR_\infty \frac{\mu_\mu}{\mu_0}\frac{e}{}\right)\left(1+\frac{m_e}{m_\mu}\right)^{-3} \times (1+\tfrac{3}{2}\alpha^2 + a_e + \epsilon_1 + \epsilon_2 + \epsilon_3 - \delta_\mu) \quad (1)$$

in which

$$a_e = \frac{\alpha}{2\pi} - 0.328\frac{\alpha^2}{\pi^2}, \quad \epsilon_1 = \alpha^2(\ln 2 - \tfrac{5}{2}),$$

$$\epsilon_2 = \frac{8\alpha^3}{3\pi}\ln\alpha\left(\ln\alpha - \ln 4 + \frac{281}{480}\right), \quad \epsilon_3 = \frac{\alpha^3}{\pi}(18.4 \pm 5),$$

$$\delta_\mu = \frac{3\alpha}{\pi}\frac{m_e m_\mu}{m_\mu^2 - m_e^2}\ln(m_\mu/m_e).$$

A value for the fine-structure constant α can be obtained by equating $\Delta\nu_{\text{theor}}$ to $\Delta\nu_{\text{expt}}$. Accepted values[3,9] are used for the constants appearing in the expression for $\Delta\nu_{\text{theor}}$. Only the value for the ratio of the magnetic moment of the muon to that of the proton, μ_μ/μ_p, requires special comment because the uncertainty with which it is known contributes significantly to the error in the determination of α. The value taken is

$$\mu_\mu/\mu_p = 3.183\,355 \pm 0.000\,048$$

(±15 ppm, 1 standard deviation).

The basic experimental measurement[10] was the ratio of the muon precession frequency in water to the proton precession frequency in water. The value quoted is the average of two values for μ_μ/μ_p. One is based on a model in which the muon and the proton in water experience the same magnetic field shielding (giving 3.183 38 ± 13 ppm). The other is based on a specific model[4] in which the shielding is different (giving 3.183 33 ± 13 ppm). The error given takes into account the error of the experiment and also the uncertainty in the shielding correction. The value obtained for α is $\alpha^{-1} = 137.0368 \pm 0.0012$ (±9 ppm, 1 standard deviation), in which the uncertainty comes principally from the uncertainty in the muon magnetic moment.

Alternatively, if one accepts a value for α, the experimental value of $\Delta\nu$ can be used to determine the muon magnetic moment (or the muon mass).[11] With the use of the ac Josephson value[12] $\alpha^{-1} = 137.0359 \pm 0.0004$ one obtains

$$\mu_\mu/\mu_p = 3.183\,314 \pm 0.000\,034$$

(11 ppm, 1 standard deviation).

From the relation

$$\frac{m_\mu}{m_e} = \frac{\mu_p}{\mu_\mu}\frac{\mu_e}{\mu_p}\frac{g_\mu}{g_e}$$

in which g_e and μ_e are the electron g value and magnetic moment, one can obtain a more precise value for the muon mass.[13] The value we obtain after inserting accepted values[3,9,14] for μ_e/μ_p and g_μ/g_e is

$$m_\mu/m_e = 206.770 \pm 0.002$$

(11 ppm, 1 standard deviation).

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DEPENDENCE OF NUCLEAR POLARIZED Fe^{57} MÖSSBAUER SPECTRA ON THE MAGNITUDE AND SIGN OF THE HYPERFINE FIELD

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The Mössbauer spectra of Fe^{57}, with the Co^{57} source in iron and palladium matrices, respectively, have been measured in a He^3/He^4 dilution refrigerator. Both the magnitude and sign of the effective field at Co^{57} can be determined from nuclear polarization effects in the spectra, without recourse to an external magnetic field. Experimental data are also presented on the Kapitza thermal boundary resistance which, together with radioactive self-heating, prevents the source from cooling below 0.15°K.

The possibility of observing nuclear polarization by using the Mössbauer effect was first suggested by Dash et al.,[1] who also made the first quantitative measurements on Co^{57} nuclei in an iron matrix.[2-4] Later experiments[5,6] in our laboratory, employing a He^3/He^4 dilution refrigerator, extended this work to considerably lower temperatures. The present paper discusses similar measurements made on Co^{57} in a Pd matrix and shows that the intensity ratios of the Mössbauer lines can give not only the magnitude but also the sign of the hyperfine field on the parent Co^{57} nucleus.

When we study the nuclear polarization of Co^{57} by the Mössbauer effect we have to take into account several nuclear levels.[2] The Co^{57} ground state with spin $I_3 = \frac{7}{2}$ (the subscript refers to the nuclear level in question) decays by electron capture to the Fe^{57} 136-keV level with spin $I_2 = \frac{5}{2}$ and mean lifetime 9×10^{-9} sec. This state decays principally by the emission of a 123-keV gamma ray to the first excited state of Fe^{57}, with $I_1 = \frac{3}{2}$ and lifetime 1.1×10^{-7} sec. The well known 14.4-keV Mössbauer gamma transition occurs between this state and the Fe^{57} ground state, $I_0 = \frac{1}{2}$.

If the degeneracy of the long-lived Co^{57} parent ground state is lifted by a negative magnetic hyperfine field, as in an iron matrix,[4] then the sublevel with magnetic quantum number $m_3 = -\frac{7}{2}$ will become preferentially populated at low enough temperatures. When the Co^{57} decays, the selection rules operate in such a way that some polarization will be preserved right through the decay chain to the 14.4-keV state of iron to give for instance more $m_1 \to m_0 = -\frac{3}{2} \to -\frac{1}{2}$ than $\frac{3}{2} \to \frac{1}{2}$ gamma transitions, i.e., the line of most negative velocity in the Mössbauer spectrum will be more intense than the corresponding positive velocity line. If H_{eff} at the parent nucleus is positive the situation is reversed.

We can assume that the nuclear spin populations of the two intermediate states 1 and 2 remain constant during the decay process because their lifetimes are short compared with estimates of the nuclear relaxation time.[2] We thus need only the nuclear magnetic moment of Co^{57} in its ground state, the appropriate Clebsch-Gordan coefficients, and the temperature for calculating the hyperfine field at the Co^{57} nucleus from the intensity ratios of the Mössbauer spectrum. Whether the most enhanced line is at the positive or negative end of the velocity spectrum gives the sign of the field. This conclusion relies on our assumption that there is no significant change