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Giant Hyperfine Anomaly between Bound Negative Muon and Rh Nucleus in Pd Metal

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The Knight shift of negative muon (μ^-Pd) in Pd metal has been determined to be $-(9.0 \pm 0.7)\%$ at $T = 11$ K, revealing an unusually large hyperfine anomaly between μ^-Pd and its equivalent isotope Rh*Pd*; $H_{\mathrm{hf}}(\mu^-\mathrm{Pd})/H_{\mathrm{hf}}(\mathrm{Rh}Pd) = 0.64 \pm 0.05$, or $\epsilon_\mu - \epsilon_N = -(36 \pm 5)\%$. Its implication is discussed in terms of the spatial distribution of the electron spin density in transition metals.

The hyperfine anomaly, the change of the hyperfine field between different nuclear states of the same isotope, reflects spatial distributions of both nuclear magnetization and electron spin density. This problem was first studied theoretically by Bohr and Weisskopf,[1] and then, rigorously by Stroke, Blin-Stoyle, and Jaccarino.[2] They considered the radial decrease of s-electron density that is probed by nuclei of finite size. Another possible cause of hyperfine anomaly is the change of $|\psi_e(0)|^2$ due to the change of the charge distribution of the probe nucleus (so-called Rosenthal-Breit-Crawford-Schawlow correction,[3] as discussed in Ref. 2). A large amount of experimental data have been explained by taking into account the nuclear magnetization distributions consistent with the nuclear wave functions, but neither the nuclear structure nor the mechanism of the hyperfine field become clear from these studies simply because the nucleus is too small to produce a large effect sensitive enough to discriminate between models. It would be dramatic, if we could find a much more extended magnetic probe to detect the electron spin density.

In this context, we paid special attention to the hyperfine field probed by bound muons.[4] Polarized negative muons that have stopped in a material immediately reach the ground state ($1s_{1/2}$) of the muonic atom and stay for a certain length of time τ_μ (τ_μ varies from 2.2 μsec in the lightest atoms to 80 nsec in heavy atoms). The average polarization is decreased to about $\frac{1}{6}$ due to the spin-orbit coupling, but is still large enough to observe spin precession (negative-muon spin rotation). The density of the bound μ^- is given by $|\psi_{1s}{}^\mu(r)|^2$. The muon wave function $\psi_{1s}{}^\mu(r)$ is, for a point nucleus, $\exp(-Zr/a_\mu)$ with $a_\mu = \hbar^2/m_\mu e^2 = 260$ fm, but, for heavy nuclei, where a_μ becomes close to the nuclear radius, it is modified due to the finite extension of nuclear charge. In either case, the bound muon is distributed largely outside the nucleus. However, when viewed from atomic electrons, the bound muon is still concentrated around the nucleus so that it should behave like an impurity nucleus of apparent charge $(Z-1)e$. Compared to its equivalent nucleus of true nuclear charge $(Z-1)e$, the bound muon has the following unique properties: (i) a peculiar charge distribution—the genuine nuclear charge Ze is surrounded by negatively charged cloud of distribution $|\psi_{1s}^{\mu}(r)|^2$, and (ii) in the case of even-even nuclei (we will only consider the case $I=0$), the magnetization is carried only by the muon spin which is distributed with the density $|\psi_{1s}^{\mu}(r)|^2$ and thus can be calculated exactly, while the nuclear magnetization is confined within the nuclear sphere. Therefore, we expect a large hyperfine anomaly which may hopefully tell us the mechanism of the hyperfine field, but such a physical observable has not been known to date.

By this new method, we attempted to study the core polarization phenomena in magnetic hyperfine fields. The origin of negative hyperfine fields in magnetic ions has been explained by Freeman and Watson[5] in terms of the induced polarization of innershell s electrons. Having such an extended magnetic probe as μ^-, we hoped to examine this mechanism on a firm experimental basis. For this purpose, the case of μ^-Pd in pure Pd metal was chosen because its nuclear counterpart, namely the Rh Knight shift in Pd metal, was known to have a very large negative value [$-(14.7\pm0.3)\%$ at 4.2 K] from a perturbed-angular-correlation measurement of ^{100}Rh in Pd metal by Rao, Matthias, and Shirley,[6] indicating that the core polarization as well as enhanced moment localization plays important roles. The relaxation time T_1 of ^{103}Rh as well as its Knight shift was measured in an NMR experiment by Narath and Weaver.[7] If no hyperfine anomaly is present, we expect that $T_1(\mu^-\text{Pd})=16$ μsec at 4 K which is much longer than $\tau_\mu=80$ nsec.

In a very early experiment, Ignatenko[8] observed no μ^- precession signal in Pd at room temperature. Later, in our previous experiment,[9] we cooled down a very pure Pd metal to 4 K, but no precession signal was observed. This result led us to a measurement of the circular polarization of muonic K x rays,[10] which showed that the muon polarization survives until the muon reaches the ground state. In the present experiment we challenged this problem again, and finally obtained precession signals, yielding a large μ^- hyperfine anomaly for the first time.

The experiment was carried out at the stopped muon channel of Clinton P. Anderson Meson Physics Facility (LAMPF). High-purity Pd wires with impurity concentration below 5 ppm were stacked together into approximate dimensions of 5 cm×2 cm×2 cm and were cooled down to 11 K in a helium-flow cryostat. A Varian 12-in. magnet with a pole gap of 5.2 cm was used to achieve a good field homogeneity; the field was monitored by an NMR gaussmeter to check the long-term field drift. The "backward" μ^- beam (80 MeV/c) was collimated to 1.9 cm in diameter and was stopped in the sample. The average stopping rate was 2×10^4 μ^-/sec. Two sets of electron telescopes, both placed at forward (downstream) directions, recorded μ-e decay time spectra. The time spectra were taken by two independent time-to-amplitude-converter–analog-to-digital-converter systems interfaced to a PDP-11 computer via a MBD-11 microprogrammed CAMAC branch driver.

Measurements were performed at 3.6 kOe and at 5.0 kOe external fields. Each time spectrum was assumed to take the form

$$N(t)=N_1\exp(-t/\tau_1)[1+A\cos(\omega t+\varphi)] +N_2\exp(-t/\tau_2)+N_3, \qquad (1)$$

where the second term represents a long-lived background from surrounding materials and the last term is a constant background. The following analysis was performed on the background-corrected data: We fixed the phase φ to be a "physical" value corresponding to the counter geometry and evaluated the asymmetry A and χ^2 value for each frequency point. To find the precession frequency, we imposed the criterion that the asymmetry of the (candidate of the) precession signal should come out with a correct sign. We also demanded consistencies between the two electron telescopes, and between the data taken at two different fields. In Fig. 1, we pre-

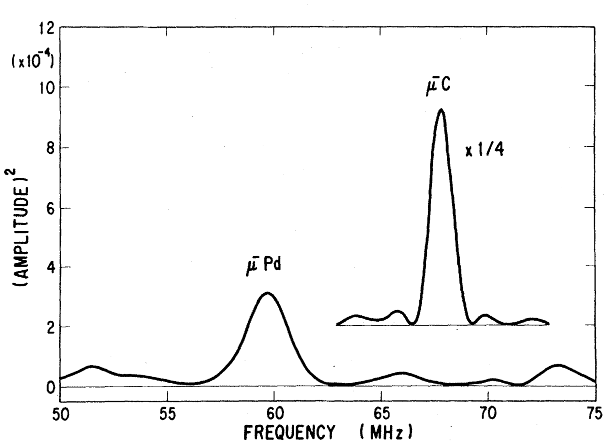

FIG. 1. Fourier power spectra, $|A|^2$ vs f of the μ^-Pd (at 11 K) and μ^-C precessions at $H_0=5.0$ kOe. The statistical fluctuations of $|A|^2$ are 0.2×10^{-4} for μ^-Pd and 0.6×10^{-4} for μ^-C.

sent the Fourier analysis of 5.0-kOe measurement. The external field at the sample position was calibrated by observing the μ^- precession in a graphite sample.

The amplitude of μ^-Pd precession was $(1.73 \pm 0.45)\%$. As shown in Fig. 1, the precession signal is statistically significant. This amplitude is much smaller than that of μ^-C, $(5.5 \pm 0.8)\%$, even after a correction is made for the natural abundance (22.2%) of ^{105}Pd [muons bound to odd-A nuclei ($I \neq 0$) are further depolarized due to the hyperfine coupling and do not contribute to the precession signal]. The reason for this small amplitude is not clear, as the relaxation time of μ^-Pd is expected to be 6 μsec at 11 K from the T_1T value for Rh*Pd*. One possible reason may be the following: Because of the preceding K x ray of about 3 MeV, the μ^-Pd 1s state is formed with a recoil energy around 50 eV, which is large enough to repel the muonic atom from its original site. This situation is somewhat similar to the final stopping stages of recoil atoms after neutron capture or nuclear reactions. As known from hyperfine-interaction studies in these cases, some significant fraction of the recoil atoms is settled down at another regular site, but some other fraction is splitted into various possible locations, thus giving rise to no precession signal (loss of orientation), since the internal field felt by a recoil nucleus depends on its location in magnetic materials, similar to the case of eqQ for $I \geqq 1$ probes. Thus, the small amplitude of μ^-Pd precession may be due to this type of loss of polarization, but the present μ^-Pd precession signal is most likely to correspond to the μ^-Pd sitting as a substitute of a Pd atom in a Pd metal, just like the Rh atom in a Rh*Pd* alloy.

After correcting each datum for the g factor of the bound μ^-, which is -0.07% for C and -2.0% for Pd less than the free-muon g factor (since the experimental values are not accurate enough,[11] we assumed the theoretical values by Ford *et al.*[12]). We finally deduced the μ^-Pd Knight shift in Pd to be $K_\mu = -(9.0 \pm 0.7)\%$ at $T = 11$ K. Compared with the Knight shift of Rh in Pd, $K(\mathrm{Rh}Pd) = -14.0\%$ at 11 K, which is obtained by interpolating the ^{100}Rh data by Rao, Matthias, and Shirley,[6] this leads to a surprisingly large hyperfine anomaly,

$$\Delta = \epsilon_\mu - \epsilon_N = \frac{K(\mu^-\mathrm{Pd}) - K(\mathrm{Rh}Pd)}{K(\mathrm{Rh}Pd)} = -(36 \pm 5)\%. \qquad (2)$$

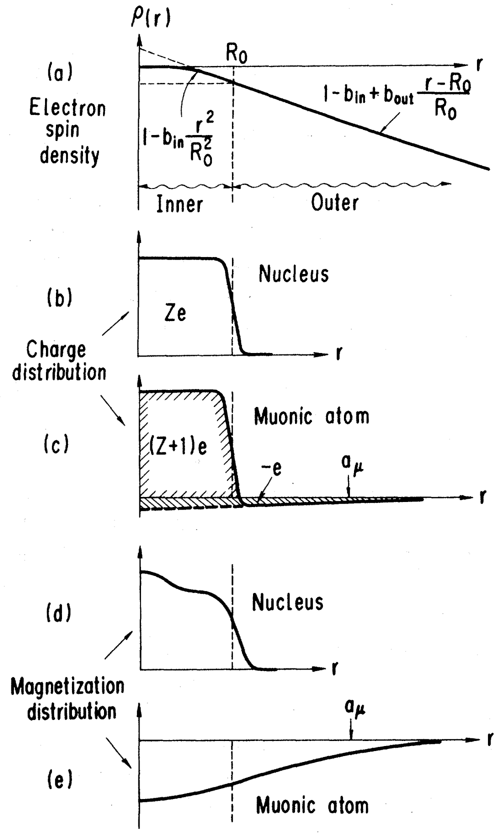

FIG. 2. The origin of the hyperfine anomaly between a nucleus of charge Ze and a muon bound to a nucleus of charge $(Z+1)e$ is schematically illustrated. The electron spin density, if it follows the s-state density, decreases quadratically inside the nucleus, and almost linearly outside the nucleus (a). The slight difference in the charge distribution between the nucleus (b) and the muonic atom (c) has little effect on the spin density, while there may exist a large hyperfine anomaly between the two since the magnetization distributions are entirely different [(d) and (e)].

In the following we will give a qualitative account of the present experiment.

The hyperfine anomaly defined with respect to the hypothetical point-nucleus value can be expressed by (neglecting the Rosenthal-Breit-Crawford-Schawlow effect)

$$\epsilon = \int_{\mathrm{probe}} m(r)[\rho(r)/\rho(0)]\, d\tau_r - 1, \qquad (3)$$

where $m(r)$ is the normalized magnetization distribution of a probe (nucleus or muonic atom) and $\rho(r)$ is the electron spin density. If the spin density is produced by a $s_{1/2}$ electron as in a free atom, $\rho(r)$ is $|\psi_s(r)|^2$ in the nonrelativistic expression, which takes the following form in the vicinity of the nucleus (see Fig. 2),

$$|\psi_s(r)|^2 = |\psi_s(0)|^2 \begin{cases} 1 - b_{\rm in}\dfrac{r^2}{R_0^{\,2}}, \quad b_{\rm in} = \dfrac{ZR_0}{a_0} \text{ for } r < R_0, & \text{(4a)} \\ 1 - b_{\rm in} - b_{\rm out}\dfrac{r-R_0}{R_0}, \quad b_{\rm out} = \dfrac{2ZR_0}{a_0} \text{ for } r > R_0, & \text{(4b)} \end{cases}$$

where R_0 is the nuclear radius, and expressions for $b_{\rm in}$ and $b_{\rm out}$ are for the case of uniform nuclear charge distribution. In the relativistic expression, $\rho(r)$ is given by $\int_r^\infty FG dr$, and the coefficients, $b_{\rm in}$ and $b_{\rm out}$, can be calculated numerically. For μ^-Pd, we obtained $b_{\rm in} = 0.72\%$ and $b_{\rm out} = 1.01\%$.

In the case of nuclear hyperfine anomalies the integration in Eq. (3) is confined within the nuclear sphere, while, in the muonic atom, $m(r)$ distributes outside the nuclear sphere so that

$$\epsilon = -b_{\rm in}\int_0^{R_0} m(r)\frac{r^2}{R_0^{\,2}}\,d\tau - b_{\rm out}\int_{R_0}^\infty m(r)\frac{r-R_0}{R_0}\,d\tau - b_{\rm in}\int_{R_0}^\infty m(r)\,d\tau. \tag{5}$$

We evaluated the integrals numerically using the $1s_{1/2}$ muonic wave function. The integrals were obtained to be $\langle (r/R_0)^2\rangle_{\rm in} = 0.086$, $\langle [(r-R_0)/R_0]\rangle_{\rm out} = 1.23$ and $\langle 1\rangle_{\rm out} = 0.84$. This result clearly shows that the electron spin density *outside* the nucleus contributes dominantly to the muonic hyperfine anomaly ($\epsilon_\mu \simeq -1.9\%$), because the muon stays outside with probability of 84%. The nuclear hyperfine anomaly is around $\epsilon_N \simeq -0.4\%$. The difference, $\Delta = \epsilon_\mu - \epsilon_N \simeq -1.5\%$, is much smaller than the observed value $\Delta = -36\%$. Then, we ask why such a large discrepancy could occur. Consideration of the muon-electron interaction, as discussed by Mallow, Desclaux, and Freeman,[13] cannot help this situation. Note that in Eq. (4) the radial gradient is $2Z/a_0$, independent of the principal quantum number. In this sense, there is no way of removing this discrepancy, unless we assume a steeper gradient of $\rho(r)$.

In magnetic ions, where the s-d interaction induces polarization of inner s-shell electrons by the d-shell electrons of the atom according to Freeman and Watson,[5] the electron spin density is due to a small difference of large $|\psi_s^{\uparrow}(r)|^2$ and $|\psi_s^{\downarrow}(r)|^2$, which behaves differently from $|\psi_s(r)|^2$ itself, as illustrated in Ref. 5. The asymptotic form of $\rho(r)$ is not known, but let us assume that $\rho(r)$ decreases linearly outside the nucleus. Then, the present experiment infers that the gradient of $\rho(r)$ is about 20 times $2Z/a_0$. This is an entirely new type of information which can be obtained only by the negative muon as a probe outside the nucleus. Whether or not this interpretation is consistent with the core polarization theory is an open question.

In the case of light nuclei, where the nuclear finite-size effect as well as the relativistic effect is unimportant for both electron and muon, we can derive a simple expression

$$\epsilon_\mu = -3(m_e/m_\mu) = -1.5\%, \tag{6}$$

irrespective of Z. Let us mention that the realistic calculation even for μ^-Pd is close to the above estimate. The nuclear hyperfine anomaly for light atoms is, of course, negligibly small. Therefore, an experimental value of ϵ_μ will lead to the gradient of $\rho(r)$ in a straightforward way. We have already observed a large negative Knight shift for μ^-Si in a weak itinerant magnet MnSi. An experiment to obtain the Knight shift of its partner, Al as a substitute for Si in MnSi, is in progress.

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