Muon spin relaxation in silicon

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A method has been developed for determining parameters describing the interaction of a positive muon in a semiconductor. Theoretical parameters characterizing the charge exchange and the orbitally bound paramagnetic state of (μ^+e^-) are determined through a comparison with experimental data on muon depolarization in silicon at $T = 290$ K. Charge-exchange frequencies have been determined for the first time.

The interactions of muons and of muonium (μ^+e^-) in semiconductors, particularly silicon, have been the subject of many studies (see the review by Patterson[1]). These are complex interactions, and many details are not yet clear. In this paper we examine experiments on muon spin relaxation in silicon in longitudinal and transverse magnetic fields. For assistance in analyzing these experiments, we offer a systematic theory of the relaxation process, which terminates in the capture of the muon in a diamagnetic compound. We focus on an n-type silicon single crystal with a resistivity of 30 $\Omega\cdot$cm and a dopant concentration $\sim 10^{14}$ cm^{-3}.

The experimental results on spin relaxation in this sample were found previously.[2] The measurements were carried out at a temperature $T = 290$ K in the JINR accelerator in Dubna.

The time evolution $N(t)$ of the number of positrons resulting from the decay $\mu^+ \to e^+$ is written in the form

$$N_\parallel(t) = N_0 e^{-t/\tau_0}(1 - c_\parallel e^{-\Lambda_\parallel t}) + B \tag{1}$$

in a longitudinal magnetic field $H_\parallel$ or in the form

$$N_\perp(t) = N_0 e^{-t/\tau_0}(1 - c_\perp e^{-\Lambda_\perp t}\cos\omega t) + B \tag{2}$$

in a transverse magnetic field $H_\perp$. Here $\tau_0 = 2.2\cdot 10^{-6}$ is the muon lifetime, $\Lambda_\parallel$ and $\Lambda_\perp$ are the relaxation rates of the muon spin, $c_\parallel$ and $c_\perp$ are experimental coefficients in the angular distribution of positrons from the decay $\mu^+ \to e^+$, B is a background, $\omega = \gamma_\mu H_\perp$ is the Larmor precession frequency of the muon spin in a field $H_\perp$, and γ_μ is the gyromagnetic ratio of the muon. The exponential behavior of the muon-spin relaxation function $P_{\rm exptl}(t)$ is confirmed by experimental data. No muon spin precession at the muonium frequency was observed in this silicon sample at $T = 290$ K. A muonium precession with an amplitude $c_\perp^{Mu} = 0.167 \pm 0.027$ was detected at a low temperature, $T = 86$ K.

Figures 1 and 2 show the experimental behavior $\Lambda_\parallel(H_\parallel)$ and $c_\parallel(H_\parallel)$ found from (1) and (2). Table I shows values of $c_\perp(H_\perp)$ and $\Lambda_\perp(H_\perp)$.

The coefficients $c_\parallel$ and $c_\perp$ measured in silicon must be compared with the asymmetry coefficient a in a material which does not cause depolarization. The value $a = 0.303 \pm 0.004$ was found in measurements of the time evolution $N_\perp(t)$ in copper. The following qualitative conclusions can be drawn from the experimental results presented above.

It can be seen from Fig. 1 that as the longitudinal field $H_\parallel$ is increased the relaxation rate $\Lambda_\parallel$ decreases, but even at $H_\parallel \approx 5$ kOe the rate $\Lambda_\parallel = 1\ \mu$s^{-1} is still significant. This $\Lambda_\parallel(H_\parallel)$ behavior can be observed only in the interaction of the muon spin with the magnetic moment of an electron in an orbitally bound paramagnetic state, i.e., in muonium (μ^+e^-).

It follows from Fig. 2 that the coefficient $c_\parallel$ is much smaller than the value of a in any field $H_\parallel$. Table I shows that the relation $c_\perp < a$ is observed in a transverse magnetic field $H_\perp$ also; this result suggests a substantial depolarization of the muon spin over an unobservably short time $\Delta t < 10$ ns. It follows from Fig. 2 that this rapid depolarization is partially suppressed by a longitudinal magnetic field $H_\parallel \lesssim 1$ kOe. It is natural to suggest that the rapid depolarization of the muon occurs in a state of short-lived muonium which forms immediately after the ionizational stopping of the muon in the silicon. It can also be seen from Fig. 2 that the coefficient $c_\parallel$ falls off monotonically with increasing field $H_\parallel$ at $H_\parallel \gtrsim 1$ kOe. Such a decrease in $c_\parallel$ could result only from the formation of a diamagnetic (chemical) compound in which a nonzero muon spin polarization is preserved. The same process—the formation of this chemical compound—is primarily responsible for the weak $\Lambda_\parallel(H_\parallel)$ dependence in longitudinal fields $H_\parallel \gtrsim 3$ kOe (Fig. 1).

Important experiments for describing the muon spin relaxation in silicon are those in a transverse magnetic field in which the muon precesses at the Larmor frequency of the

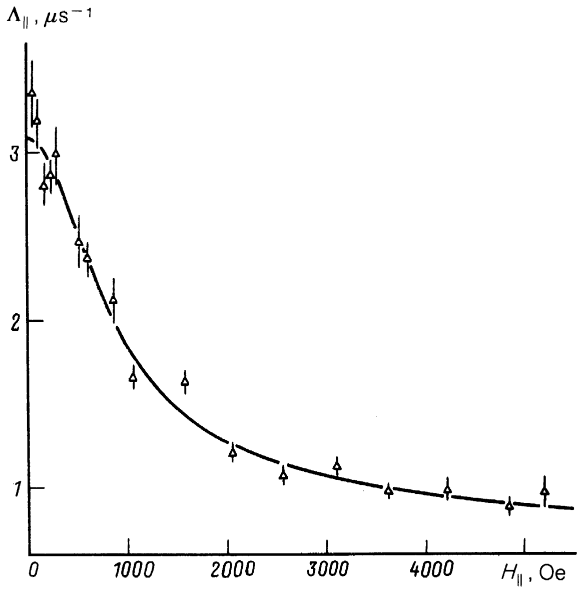

FIG. 1. Relaxation rate of the muon spin, $\Lambda_\parallel$, versus the longitudinal magnetic field $H_\parallel$. The solid line shows the theoretical $\Lambda_\parallel^{\rm theo}(H_\parallel)$ dependence [see (13)].

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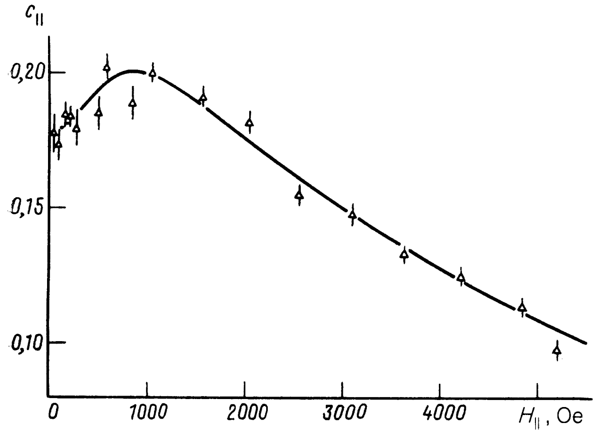

FIG. 2. The $c_{\parallel}(H_{\parallel})$ dependence, i.e., the asymmetry coefficient of the angular distribution of positrons from the decay $\mu^+ \to e^+$ versus the longitudinal field $H_{\parallel}$. The solid line is the theoretical $c_{\parallel}^{\text{theo}}(H_{\parallel})$ dependence [see (12)].

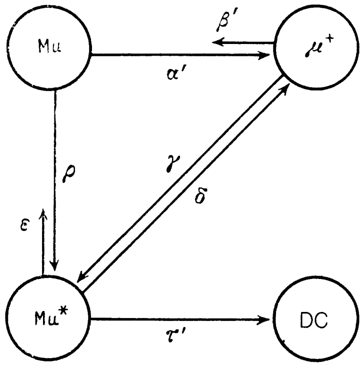

FIG. 3. Schematic diagram of the relaxation of the muon spin in silicon. *Mu*—normal muonium; *Mu**—anomalous muonium; μ^+—free muon; DC—diamagnetic (chemical) compound.

free muon. It follows that, as we have already concluded, the paramagnetic state of (μ^+e^-) is unstable and rapidly undergoes a transition to a state of a free muon:

$$(\mu^+e^-)\rightleftharpoons\mu^+. \tag{3}$$

It is the precession of the free muon which we observe in a transverse magnetic field. On the other hand, the depolarization of a muon in longitudinal and transverse magnetic fields results from the effect of these fields on the short-lived muonium which is periodically formed as a result of the fast charge-exchange processes represented by (3). A longitudinal field suppresses the depolarization of the muon in muonium, while a transverse field intensifies it. The weak dependence of $c_\perp$ and $\Lambda_\perp$ on a transverse field $H_\perp$ which follows from Table I, combined with the strong dependence of $\Lambda_{\parallel}$ on a longitudinal field $H_{\parallel}$ of the same strength, $H_{\parallel} \lesssim 1$ kOe (Fig. 1), means that transverse fields $H_\perp = 40$–700 Oe cause an essentially complete depolarization of the muon in muonium (μ^+e^-). This effect could occur only if the short—lived muonium nevertheless lived long enough for the muon to undergo depolarization in fields $H_\perp > 40$ Oe.

We turn now to a quantitative description of the muon spin relaxation in silicon. In accordance with the discussion above, we show in Fig. 3 a diagram of the relaxation process which precedes the capture of the muon in a diamagnetic (chemical) compound. Here *Mu* is a short-lived paramagnetic compound which we will assume to be normal muonium; *Mu** is anomalous muonium; μ^+ is a free muon; and DC is a stable diamagnetic (chemical) compound. The arrows show transitions between states. The short and long arrows are meant to represent slow and fast transitions. The Greek letters $\alpha', \beta', \gamma, \delta, \rho$, and ε are the probabilities for the corresponding transitions; and τ' is the lifetime of *Mu** with respect to conversion into the chemical compound.

The transition $\mu^+ \rightleftarrows Mu^*$ depicted in Fig. 3 was not mentioned in the qualitative description of the experiment. In a quantitative calculation, however, it becomes necessary to incorporate the transitions γ and δ, which determine one more process by which the muon spin relaxes. We will be describing this process by means of a magnetic-field-dependent parameter Λ^*, which is the relaxation rate of the muon spin in the fast charge-exchange processes $\mu^+ \rightleftarrows Mu^*$. The parameters $\alpha', \beta', \rho, \varepsilon$, and τ' in Fig. 3 can then be replaced by corresponding effective values α and β, which describe the charge exchange of *Mu* in an interaction with a complex $(\mu^+ \rightleftarrows Mu^*)$, and by the lifetime (τ) of this complex with respect to the incorporation of the muon in a chemical compound. Here α and β are the probabilities for a transition of the muon from state *Mu* to the state $(\mu^+ \rightleftarrows Mu^*)$ and vice versa, respectively.

The relations $\alpha' \gg \beta'$ and $\rho \gg \varepsilon$ (or, equivalently, $\alpha \gg \beta$) indicated in Fig. 3 follow from the relatively short lifetime Δt found for the *Mu* state in a qualitative analysis. The high probability for the transitions γ and δ corresponds to a low relaxation rate Λ^* of the muon spin in the state $(\mu^+ \rightleftarrows Mu^*)$ and to a muon precession frequency for the muon spin in a transverse magnetic field $H_\perp$. We derive below a method for determining the frequencies α, β, γ, and δ experimentally. That the transitions (charge exchanges) shown in Fig. 3 actually occur has been established qualitatively in several experiments (see the review by Patterson[1]).

We denote by P_{Mu}, P_{Mu^*}, P_{μ^+}, and P_{chm} the components of the polarization of muons in the states *Mu*, *Mu**, μ^+, and the diamagnetic compound, respectively. By "component of the polarization" we mean here the product of the polarization of the muon in the given state and the probability that the muon will be in this state. The total polarization of the muon is then written in the form

$$P(t) = P_{Mu} + P_{Mu^*} + P_{\mu^+} + P_{\text{chm}}. \tag{4}$$

The corresponding relaxation process, i.e., the $P(t)$ dependence, is described by the equations

$$P_\Sigma(t) = (1-W)\exp\left[-\left(\beta+\Lambda^*+\frac{1}{\tau}\right)t\right] + \alpha\int_0^t P_{Mu}(t') \times\exp\left[-\left(\beta+\Lambda^*+\frac{1}{\tau}\right)(t-t')\right]dt', \tag{5}$$

TABLE I. The parameters $c_\perp$ and $\Lambda_\perp$ in a transverse field $H_\perp$.

$H_\perp$, Oe	$c_\perp$	$\Lambda_\perp$, μs^{-1}
44	0,135±0,009	4,5±0,7
200	0,154±0,019	4,5±0,7
670	0,139±0,017	4,1±0,6

$$P_{Mu}(t)=WP_1(t)e^{-\alpha t}+\beta\int_0^t P_\Sigma(t')P_1(t-t')\exp[-\alpha(t-t')]dt', \tag{6}$$

$$P_{\text{chm}}(t)=\frac{1}{\tau}\int_0^t P_\Sigma(t')dt'. \tag{7}$$

Here $P_\Sigma = P_{\mu^+} + P_{Mu^*}$; $P_1(\nu,H_0,H,t)$ is the polarization of the muon in state Mu which has persisted to the time t; W is the probability for the formation of Mu at the initial time ($t=0$); ν is the frequency of the electron spin flip in Mu; and H_0 is the hyperfine field at the electron of the muonium.

The structure of Eqs. (5)–(7) is understandable. A similar system of equations was studied in Ref. 3. They were solved by the standard Laplace method. The relaxation function $P(t)$ [see (4)] found in the course of the solution for the case $\alpha\gg\beta$, under consideration here and under the condition $t\gg 1/\omega_0$, where $\omega_0\approx 10^{10}$ s^{-1} is the hyperfine splitting frequency of normal muonium, can be written in the form

$$P(t)=P_\infty+(P_0-P_\infty)e^{-\Lambda t}, \tag{8}$$

$$\Lambda=\left[(1-\mathcal{P}_1)\beta+\Lambda^*+\frac{1}{\tau}\right]\Big/\left(1+\frac{\beta}{\alpha}\mathcal{P}_1-\beta\frac{\partial\mathcal{P}_1}{\partial\alpha}\right),$$

$$\mathcal{P}_1=\alpha\int_0^\infty P_1(t)e^{-\alpha t}dt, \tag{9}$$

$$P_0=W\mathcal{P}_1+(1-W)+\frac{W\mathcal{P}_1}{\alpha}\left[(1-\mathcal{P}_1)\beta+\Lambda^*+\frac{1}{\tau}\right]$$
$$+\frac{\partial\mathcal{P}_1}{\partial\alpha}\left\{\beta-W\left[2\beta(1-\mathcal{P}_1)+\Lambda^*+\frac{1}{\tau}\right]\right\}, \tag{10}$$

$$P_\infty=\frac{W\mathcal{P}_1+(1-W)}{\tau[(1-\mathcal{P}_1)\beta+\Lambda^*+1/\tau]}. \tag{11}$$

Expressions for $\Lambda_\parallel^*$ and $\Lambda_\perp^*$ in longitudinal and transverse magnetic fields are[4]

$$\Lambda_\parallel^*=\frac{\Lambda_0^*(1+fh^2)}{1+h^2},\qquad \Lambda_\perp^*=\frac{\Lambda_0^*}{2}\left(1+\frac{1+fh^2}{1+h^2}\right),$$

where, with $\delta\gg\omega_\perp^*$,

$$\Lambda_0^*=\frac{(\omega_\perp^*)^2\gamma q}{2\delta(\gamma+\delta)},\qquad h=\frac{\gamma_e H}{\delta},\qquad f=\frac{b^2}{27q},$$

$$q=1-\frac{b}{3}(2-b),$$

$$b=\frac{\omega_\perp^*-\omega_\parallel^*}{\omega_\perp^*}=0.84;$$

$\omega_\perp^*=2\pi\cdot 90\ \mu$s^{-1} and $\omega_\parallel^*=2\pi\cdot 14\ \mu$s^{-1} are the hyperfine splitting frequencies of anomalous (anisotropic) muon at $T=290$ K (Ref. 5); and γ_e is the gyromagnetic ratio of the electron. Expressions for $P_1(H)$ and $\mathcal{P}_1(H)$ in longitudinal ($H=H_\parallel$) and transverse ($H=H_\perp$) magnetic fields are given in Ref. 3.

Substituting (8) into (1), we find an expression for the theoretical asymmetry coefficient $c_\parallel^{\text{theo}}(H_\parallel)$ in a longitudinal magnetic field $H_\parallel$:

$$c_\parallel^{\text{theo}}=\frac{a[P_0(H_\parallel)-P_\infty(H_\parallel)]}{1-aP_\infty(H_\parallel)}, \tag{12}$$

where $P_0(H_\parallel)$ and $P_\infty(H_\parallel)$ are given by (10) and (11) in a field $H_\parallel$. Expression (9) is a theoretical expression for $\Lambda_\parallel^{\text{theo}}$; here $\mathcal{P}_1=\mathcal{P}_1(H_\parallel)$, i.e.,

$$\Lambda_\parallel^{\text{theo}}=\Lambda(H_\parallel). \tag{13}$$

In a transverse magnetic field $H_\perp$, the measured quantity $c_\perp$ is equal to the precession amplitude of the muon spin at $t=0$. Substituting expression (8) for $P(t)$ into (2), we find then

$$c_\perp^{\text{theo}}=aP_0(H_\perp). \tag{14}$$

In determining the functional $\Lambda_\perp^{\text{theo}}(H_\perp)$ dependence we should allow for the circumstance that the decay of the precession amplitude $P(t)$ in (8) is not exponential. Under the condition $P_\infty(H_\perp)\ll P_0(H_\perp)-P_\infty(H_\perp)$ which can be satisfied in experiment, however, the corresponding correction is small and can be dealt with in the following way:

$$\Lambda_\perp^{\text{theo}}=\Lambda(H_\perp)\left[1-\frac{P_\infty(H_\perp)}{P_0(H_\perp)}\right]. \tag{15}$$

The process described above for the relaxation of the muon spin in silicon is not the only one possible. In addition to the first version of the relaxation process discussed above (Fig. 3), there is a second, in which Mu rather than Mu^* enters in the chemical compound. The integral equations describing this second version of the relaxation are

$$\bar{P}_\Sigma(t)=(1-W)\exp[-(\beta+\Lambda^*)t]$$
$$+\alpha\int_0^t \bar{P}_{Mu}(t')\exp[-(\beta+\Lambda^*)(t-t')]dt', \tag{16}$$

$$\bar{P}_{Mu}(t)=WP_1(t)\exp\left[-\left(\alpha+\frac{1}{\tau}\right)t\right]+\beta\int_0^t \bar{P}_\Sigma(t')P_1(t-t')$$
$$\times\exp\left[-\left(\alpha+\frac{1}{\tau}\right)(t-t')\right]dt', \tag{17}$$

$$\bar{P}_{\text{chm}}(t)=\frac{1}{\tau}\int_0^t \bar{P}_{Mu}(t')dt'. \tag{18}$$

The solution of Eqs. (16)–(18) for the resultant polarization $\bar{P}(t)$,

$$\bar{P}(t)=\bar{P}_{Mu}+\bar{P}_{Mu^*}+\bar{P}_{\mu^+}+\bar{P}_{\text{chm}},$$

is of the same form as solution (8) for $P(t)$:

$$\bar{P}(t)=\bar{P}_\infty+(\bar{P}_0-\bar{P}_\infty)\exp(-\bar{\Lambda}t). \tag{19}$$

Here

$$\bar{P}_\infty=\frac{\mathcal{P}_1[W(\beta+\Lambda^*)+\beta(1-W)]}{\tau[(\alpha+1/\tau)(\beta+\Lambda^*)-\alpha\beta\mathcal{P}_1]}, \tag{20}$$

$$\bar{P}_0=W\mathcal{P}_1+(1-W)+\frac{W\tau\mathcal{P}_1}{\alpha\tau+1}\left[\beta(1-\mathcal{P}_1)-\frac{\alpha\beta\tau}{(\alpha\tau+1)^2}+\Lambda^*\right]$$
$$+\frac{\partial\mathcal{P}_1}{\partial\alpha}\left\{\beta-W\left[2\beta+\Lambda^*-\frac{2\alpha\beta\tau\mathcal{P}_1}{(\alpha\tau+1)^2}(\alpha\tau+2)\right]\right\}, \tag{21}$$

$$\bar{\Lambda}=\left[\frac{\beta}{\alpha\tau+1}+\Lambda^*+\frac{\alpha\beta\tau}{\alpha\tau+1}(1-\mathcal{P}_1)\right]\Big/\left(1+\frac{\beta}{\alpha}\mathcal{P}_1-\beta\frac{\partial\mathcal{P}_1}{\partial\alpha}\right). \tag{22}$$

In a comparison of the theoretical relaxation function $\bar{P}(t)$ with the experimental one we should use Eqs. (12)–(15) and expressions (20)–(22) for $\bar{P}_\infty$, $\bar{P}_0$, and $\bar{\Lambda}$.

Let us compare theoretical expressions (8) for $P(t)$ and

TABLE II. Parameters of the relaxation functions $P(t)$ and $\overline{P}(t)$ for versions 1 and 2 of the incorporation of a muon in a chemical compound, specifically from the states *Mu** and *Mu*, respectively.

Parameter	Version 1 $P(t)$	Version 2 $\overline{P}(t)$
χ^2	42	53
H_0, Oe	731±80	780±130
ν, μs^{-1}	14±13	3±4
α, μs^{-1}	65^{+60}_{-25}	181±35
β, μs^{-1}	3,4±0,2	4,3±0,4
W	0,56±0,03	0,64±0,03
$1/\tau$, μs^{-1}	0,66±0,03	30±6
Λ_0^*, μs^{-1}	0,46±0,12	0,59±0,22
γ, μs^{-1}	(3,5±0,6)·10⁴	(3,6±0,6)·10⁴
δ, μs^{-1}	$\left(7^{+2}_{-1}\right)\cdot 10^4$	(7±2)·10⁴
γ/δ	0,47±0,04	0,56±0,11

(19) for $\overline{P}(t)$ with the experimental results (1) and (2). Figures 1 and 2 show the experimental and corresponding theoretical behavior of the relaxation rate and the asymmetry coefficient as functions of the longitudinal magnetic field $H_{\parallel}$ for the case in which the muon goes into a chemical compound from the state *Mu** or, equivalently, from the state $(\mu^+ \rightleftarrows Mu^*)$.

Figures 1 and 2 reveal a good agreement between theory and experiment and thus support theoretical model. The relaxation function $\overline{P}(t)$ also agrees well with experimental data. Comparison of theoretical values (12)–(15) of $c_{\parallel}^{\text{theo}}(H_{\parallel})$, $\Lambda_{\parallel}^{\text{theo}}(H_{\parallel})$, $c_{\perp}^{\text{theo}}(H_{\perp})$, and $\Lambda_{\perp}^{\text{theo}}(H_{\perp})$ for the relaxation functions $P(t)$ and $\overline{P}(t)$ with the corresponding experimental behavior (Figs. 1 and 2 and Table I) yielded the parameters which describe the functions $P(t)$ and $\overline{P}(t)$. These parameter values are listed in Table II.

The parameter errors in Table II are the standard deviations; mutual correlations in all the parameters shown have been considered.

It can be seen from Table II that the conclusion $\alpha \gg \beta$ which was reached in the qualitative analysis of the experimental data is supported by a quantitative calculation. It also follows from Table II that the two versions of the relaxation process are described by approximately the same parameter values. The relatively high rate at which the muon goes into the chemical compound from the state *Mu*, i.e., the rate in the second version of the relaxation process, is a consequence of the short time spent by the muon in state *Mu*. The absolute rate at which the muon goes into the chemical compound in the second version,

$$\left(\frac{1}{\tau}\right)_{\text{abs}} = \frac{\beta}{\alpha+1/\tau}\frac{1}{\tau} = 0{,}61\pm0{,}02$$

is the same as the rate of the chemical reaction of the muon from the state *Mu**.

This result shows that the experimentally determined rate $[(1/\tau)_{\text{abs}}]$ at which the muon undergoes a transition into a diamagnetic state does not depend on the particular state, *Mu** or *Mu*, from which the transition is assumed to occur. The values found for the parameters $H_0, \nu, \beta, W, \Lambda_0^*$, γ, and δ in this experiment are also independent of the particular version of the chemical process. A comparison of the parameter values for versions 1 and 2 on the basis of the χ^2 test (41 values of experimental quantities with 9 theoretical parameters determined) shows that the first version of the relaxation function, $P(t)$, is statistically more reliable. This is the version in which the muon goes into the chemical compound from the state *Mu**. The value $H_0 = 731 \pm 80$ Oe found for this version agrees with the value $H_0 = 690$ Oe found in measurements of the hyperfine-splitting frequency of muonium in ultrapure silicon at $T = 290$ K (Ref. 6). The probability W listed in Table II for the formation of *Mu* in silicon at $T = 290$ K agrees with the value $W = 0.61 \pm 0.08$ found through direct measurements at a low temperature (see the review by Patterson[1]).

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