

Muon acoustic resonance in semiconductors and dielectrics

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Muonium spin-phonon interaction is studied in semiconductors and dielectrics under conditions of muon acoustic spin resonance (μ ASR). General expressions are derived for the muon spin behavior under periodic perturbation near the resonance. It is shown that under these conditions the periodic perturbation manifests itself in the form of characteristic oscillations of the longitudinal polarization or of minima of the average polarization. The oscillation frequency or minimum depth is given by the interaction magnitude. The formulas obtained are applied to μ ASR-experiments with isotropic Mu in diamond-structure crystals. It is shown that μ ASR-experiments yield all the constants of muonium spin-phonon interaction.

The acoustic magnetic resonance is used extensively in solid state studies. However in the acoustic nuclear magnetic resonance (ANMR) and acoustic electron spin resonance (AESR) methods the independent constants of spin-phonon interaction of a paramagnetic center are obtained, as a rule, indirectly by analysis of the acoustic wave absorption coefficient or of the ESR or NMR signal saturation (see, e.g., Refs. 1 and 2). The μ SR method in its traditional version, implying the acoustic wave excitation, yields directly the spin-phonon interaction constants in crystals where a muonic atom is formed. The possibilities for the investigation of ferromagnets by the muon acoustic resonance (μ ASR) are analyzed in Refs. 3 and 4.

First, consider the muon spin polarization under periodic perturbation of frequency ω . The Hamiltonian of the system has the form

$$H(t) = H_0 + V(t), \quad (1)$$

where H_0 is the Hamiltonian of an unperturbed system (a muon in the lattice, muonium, etc.), and $V(t)$ is a periodic perturbation of frequency ω .

It is convenient to write the polarization in the interaction representation:

$$P_i(t) = \text{Sp } \sigma_i(t) \rho_I(t). \quad (2)$$

Here

$$\sigma_i(t) = \exp(i\hbar^{-1}H_0t) \sigma_i \exp(-i\hbar^{-1}H_0t) \quad (3)$$

is the muon spin operator in the interaction representation. The density matrix satisfies the well-known equation

$$\dot{\rho}_I(t) = -i\hbar^{-1}[V_I(t), \rho_I(t)], \quad (4)$$

where

$$V_I(t) = \exp(i\hbar^{-1}H_0t) V(t) \exp(-i\hbar^{-1}H_0t) \quad (5)$$

is the perturbation operator in the interaction representation.

The problem of quantum-system behavior under the action of periodic perturbations is well-known (see, e.g., Ref. 5). We, however, have to find the spin polarization in such a system. Consider the case when H_0 has a discrete nondegenerate spectrum. Then, in the interaction representation, the arbitrary periodic perturbation operator

$$V(t) = V \exp(-i\omega t) + \text{H.c.} \quad (6)$$

has the following form in the basis of eigenfunctions of the Hamiltonian H_0 :

$$V_I(t) = \sum_{k,n} \{ \langle n|V|k\rangle \exp[i(\omega_{nk} - \omega)t] + \langle k|V|n\rangle \exp[-i(\omega + \omega_{nk})t] \}. \quad (7)$$

Here $\hbar\omega_{nk} = \varepsilon_n - \varepsilon_k$; $|n\rangle$, $|k\rangle$ and ε_n , ε_k are the eigenfunctions and eigenvalues of H_0 respectively. In what follows we assume that $\varepsilon_n > \varepsilon_k$.

As is well known,⁵ if for some levels the resonance condition holds, $\omega_{nk} \approx \omega$ ($|\omega_{nk} - \omega|/\omega \ll 1$), the problem reduces to the two-level one. A two-level system is conveniently described by an effective spin $\tau = \frac{1}{2}$. The effective spin operator $\hat{\tau}_i$ is introduced as follows:

$$\begin{aligned} \hat{\tau}_z|n\rangle &= \frac{1}{2}|n\rangle, \quad \hat{\tau}_z|k\rangle = -\frac{1}{2}|k\rangle, \quad \hat{\tau}_+|n\rangle = 0, \\ \hat{\tau}_+|k\rangle &= |n\rangle, \quad \hat{\tau}_+ = \hat{\tau}_x + i\hat{\tau}_y, \quad \hat{\tau}_- = (\hat{\tau}_+)^* \end{aligned} \quad (8)$$

Naturally, the operators $\hat{\tau}_i$ satisfy the well-known commutation relations:

$$[\hat{\tau}_z, \hat{\tau}_\pm] = \pm \hat{\tau}_\pm, \quad [\hat{\tau}_+, \hat{\tau}_-] = 2\hat{\tau}_z. \quad (9)$$

In the space of two states, $|n\rangle$ and $|k\rangle$, the operator (7) has the form

$$V_I(t) = \hat{\tau}_+ A \exp(i\delta t) + \hat{\tau}_- A^* \exp(-i\delta t), \quad (10)$$

where $A = \langle n|V|k\rangle$ and $\delta = \omega_{nk} - \omega$. In the space of the remaining states the perturbation operator is assumed to be zero ($\langle m|V|l\rangle = 0$), since we study the system under conditions close to resonance. Accordingly, the density matrix elements in the interaction representation ρ_{nn} , ρ_{kk} and $\rho_{nk} = \rho_{kn}^*$ are changed by the perturbation (10), while the others remain constant.

The density matrix at the initial time can be written as

$$\rho(0) = \rho_\mu(0) \rho_s(0), \quad (11)$$

where

$$\rho_\mu(0) = \frac{1}{2}(1 + \sigma \mathbf{P}_0) \quad (12)$$

is the muon density matrix with the initial polarization \mathbf{P}_0 and $\rho_s(0)$ is the density matrix of the remaining part of the system. Let the z axis be parallel to \mathbf{P}_0 . Then the "resonance" part of the density matrix, $\rho_I(n,k;t)$, can be represented as

$$\rho_I(n, k; t) = \frac{1}{2} \{ 1 + P_0 [\frac{1}{2} (\sigma_{nn} + \sigma_{kk}) + (\sigma_{nn} - \sigma_{kk}) \tau_z(t) + \sigma_{nk} \tau_+(t) + \sigma_{kn} \tau_-(t)] \}. \quad (13)$$

Here $\sigma_{nn}, \sigma_{kk}, \sigma_{nk} = \sigma_{kn}^*$ are the matrix elements of the operator σ_z in the space of the states $|n\rangle$ and $|k\rangle$, and $\tau_z(t)$ and $\tau_{\pm}(t)$ are the effective spin operators in the interaction representation. As is well-known (see, e.g., Ref. 5), these operators obey the equation

$$\dot{\tau}_i(t) = -i\hbar^{-1} [V_I(t), \tau_i(t)]. \quad (14)$$

The formal solution of Eq. (14) can be written in the form of the iteration series

$$\begin{aligned} \tau_i(t) &= \hat{\tau}_i + (-i/\hbar) \int_0^t dt_1 [V_I(t_1), \hat{\tau}_i] \\ &+ (-i/\hbar)^2 \int_0^t dt_1 \int_0^{t_1} dt_2 [V_I(t_1), [V_I(t_2), \hat{\tau}_i]] + \dots \end{aligned} \quad (15)$$

For the spin $\frac{1}{2}$ the series can be summed.

The properties of two-level systems have been analyzed at length in quantum radiophysics (see, e.g., Ref. 6). Therefore, omitting the intermediate calculations, we give the final result:

$$\tau_z(t) = f(t) \hat{\tau}_z + iA f_+(t) \hat{\tau}_+ - iA^* f_-(t) \hat{\tau}_-. \quad (16)$$

Here

$$\begin{aligned} f(t) &= \Omega^{-2} (4|A|^2 \cos \Omega t + \delta^2), \quad \Omega^2 = 4|A|^2 + \delta^2, \\ f_{\pm}(t) &= \int_0^t dt' f(t') \exp(\pm i\delta t'). \end{aligned}$$

For the two other operators we have

$$\tau_{\pm}(t) = [1 + AF_{\pm}(t)] \hat{\tau}_{\pm} - A^* F_{\mp}(t) \hat{\tau}_{\mp} - iF(t) \hat{\tau}_z, \quad (17)$$

where

$$\begin{aligned} F(t) &= (2A^*/\Omega) [i(\delta/\Omega) (1 - \cos \Omega t) - \sin \Omega t], \\ F_{\pm}(t) &= \int_0^t dt' F(t') \exp(\pm i\delta t'), \end{aligned}$$

and, respectively,

$$\tau_{-}(t) = [1 + A^* F_{+}(t)] \hat{\tau}_{-} - AF_{-}(t) \hat{\tau}_{+} + iF(t) \hat{\tau}_z. \quad (18)$$

Using the formulas (13), (16)–(18), one can write down the muon spin polarization for arbitrary periodic perturbation under resonance conditions. The effect is most clearly observed in the form of longitudinal polarization oscillations of characteristic frequency Ω .

The transverse muon polarization in the Mu atom, precessing as a rule with many frequencies, becomes even more complicated, when a periodic perturbation is switched on. However, if the transverse polarization contains a conserved component, as for example in the anisotropic Mu* atom (Ref. 7), the oscillations of frequency Ω will also appear at resonance. Therefore, we will consider in what follows only the longitudinal polarization $P_{\parallel}(t)$. It can be written as

$$P_{\parallel}(t) = P_{\parallel}^{(0)}(t) + \delta P_{\parallel}(t), \quad (19)$$

where $P_{\parallel}^{(0)}(t)$ is the polarization in the absence of perturba-

tion, and $\delta P_{\parallel}(t)$ is the resonant correction determined by a part of the density matrix (13):

$$\delta P_{\parallel}(t) = \text{Sp } \sigma_z(n, k; t) (\rho_I(n, k; t) - \rho_I(n, k; 0)). \quad (20)$$

Introducing the notations

$$\sigma_{nk} = |\sigma_{nk}| e^{i\varphi}, \quad \sigma_{nk}(t) = \sigma_{nk} e^{i\omega_{nk}t}, \quad A = |A| e^{i\alpha}, \quad (21)$$

and performing long but simple calculations, we find the correction to the polarization, which has a rather cumbersome form

$$\begin{aligned} \delta P_{\parallel}(t) &= |\sigma_{nk}|^2 (\cos \Omega t \cos \omega t - \cos \omega_{nk} t) \\ &+ (1 - \cos \Omega t) \frac{|A|}{\Omega} \left\{ 2 |\sigma_{nk}|^2 \frac{|A|}{\Omega} [\cos \omega t + \cos(\omega t + 2(\varphi - \alpha))] \right. \\ &+ (\sigma_{nn} - \sigma_{kk}) |\sigma_{nk}| \frac{\delta}{\Omega} [\cos(\omega t + \varphi - \alpha) \\ &\left. + \cos(\varphi - \alpha)] - \frac{|A|}{\Omega} (\sigma_{nn}^2 - \sigma_{kk}^2) \right\} \\ &- \sin \Omega t \left\{ (\sigma_{nn} - \sigma_{kk}) |\sigma_{nk}| \frac{|A|}{\Omega} [\sin(\omega t + \varphi - \alpha) + \sin(\varphi - \alpha)] \right. \\ &\left. + \frac{\delta}{\Omega} |\sigma_{nk}|^2 \sin \omega t \right\}. \end{aligned} \quad (22)$$

The formula (22) is substantially simplified if the frequency $\omega \sim 10^9 - 10^{10} \text{ s}^{-1}$ is too high to be resolved in the experiment:

$$\begin{aligned} \overline{\delta P_{\parallel}(t)} &= - (1 - \cos \Omega t) (|A|/\Omega) (\sigma_{nn} - \sigma_{kk}) [(|A|/\Omega) (\sigma_{nn} - \sigma_{kk}) \\ &- (\delta |\sigma_{nk}|/\Omega) \cos(\varphi - \alpha)] \\ &- \sin \Omega t (|A|/\Omega) (\sigma_{nn} - \sigma_{kk}) |\sigma_{nk}| \sin(\varphi - \alpha). \end{aligned} \quad (23)$$

The resonance is observed most effectively when the external field dependence of the average longitudinal polarization is studied. If the depolarization occurs at a low rate $\lambda \ll \omega_{nk}$, the average polarization is

$$\langle P_{\parallel} \rangle = \frac{1}{\tau_{\mu}} \int_0^{\infty} P_{\parallel}(t) \exp(-\Lambda t) dt, \quad (24)$$

where $\tau_{\mu} = 2.2 \cdot 10^{-6} \text{ s}$ is the muon lifetime and $\Lambda = \lambda + \tau_{\mu}^{-1}$. Since $\omega_{nk} \gg \Lambda$, only the conserved components of $P_{\parallel}^{(0)}$ and slowly oscillating terms of the correction $\delta P_{\parallel}(t)$ will contribute to $\langle P_{\parallel} \rangle$. Thus, we have

$$\begin{aligned} \langle \delta P_{\parallel} \rangle &\approx - \frac{1}{\Lambda \tau_{\mu}} \frac{|A|^2}{\Omega^2} (\sigma_{nn}^2 - \sigma_{kk}^2) \\ &\times \left[1 - \frac{\delta}{|A|} \frac{|\sigma_{nk}| \cos(\varphi - \alpha)}{(\sigma_{nn} + \sigma_{kk})} \right] \frac{\Omega^2}{\Omega^2 + \Lambda^2}. \end{aligned} \quad (25)$$

As is seen, at exact resonance a characteristic minimum of $\langle P_{\parallel}(B) \rangle$ appears, whose depth is

$$\langle \delta P_{\parallel} \rangle_{\text{res}} = - \frac{\sigma_{nn}^2 - \sigma_{kk}^2}{\Lambda \tau_{\mu}} \frac{|A|^2}{\Lambda^2 + 4|A|^2}. \quad (26)$$

The characteristic behavior of $\langle P_{\parallel}(B) \rangle$ under periodic perturbation is shown in Fig. 1. The interaction value is determined by the oscillation frequency in (23) or by the depth of the minimum.

The Hamiltonian of the spin-phonon interaction of the Mu atom in a crystal does not differ from the corresponding Hamiltonian of a paramagnetic impurity of spin $S = \frac{1}{2}$. The spin-phonon interaction of the Mu atom with the lattice is

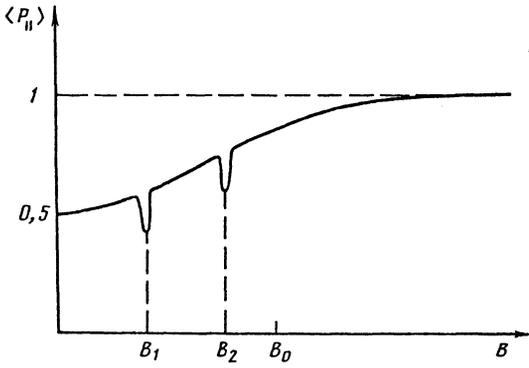


FIG. 1. Characteristic dependence of $\langle P_{\parallel} \rangle$ vs the magnetic field.

determined by the crystal field modulation in the interstice where the muonium is situated. The crystal field modulation, which decreases the local symmetry, leads to anisotropic corrections to the hyperfine-interaction constant and the g -factor of the Mu atom:

$$V_{s.ph.} = \hbar \delta \Omega_{ik} \sigma_i \sigma_k^{\mu} + \mu_0 \delta g_{ik} B_i \sigma_k^{\mu}. \quad (27)$$

The phenomenological second-rank tensors $\delta \Omega_{ik}$ and δg_{ik} are given by the crystal field gradients and are proportional to the lattice deformations:¹

$$\hbar \delta \Omega_{ik} = Z_{iklm} u_{lm}, \quad \delta g_{ik} = F_{iklm} u_{lm}. \quad (28)$$

Here Z_{iklm} and F_{iklm} are the coupling tensors and u_{lm} is the lattice deformation tensor. In the field of a standing acoustic wave the displacement vector has the form

$$\mathbf{u} = \mathbf{e} u_0 \sin(\mathbf{k}\mathbf{r} - \omega t), \quad (29)$$

where u_0 , \mathbf{e} and \mathbf{k} are the acoustic wave amplitude, polarization, and wave vector respectively. The deformation tensor

$$u_{lm} = (1/2) (\partial u_l / \partial x_m + \partial u_m / \partial x_l)$$

for the wave (29) is

$$u_{lm} = (u_0/2) (e_l k_m + e_m k_l) \cos(\mathbf{k}\mathbf{r} - \omega t). \quad (30)$$

Thus, the muon spin polarization in the presence of the perturbation (27) is determined by the formulas (22), (23), (25), and (26). Note that, as follows from (29), the perturbation phase depends on \mathbf{r} :

$$\alpha = \alpha_0 + \mathbf{k}\mathbf{r},$$

therefore the muon ensemble polarization is determined by averaging Eqs. (22) and (25) over the sample volume. However, as seen from Eqs. (23) and (26), at exact resonance the polarization, as well as the minimum depth, are independent of α .

Let us analyze the possibilities of the μ ASR-method for the simplest case of isotropic muonium.

The coupling tensors Z and F are symmetric with respect to the index permutation in the first and second pairs, i.e.,

$$Z_{iklm} = Z_{kilm} = Z_{ikml}.$$

In the physical acoustics one uses the Voigt notation

$$xx \rightarrow 1, \quad yy \rightarrow 2, \quad zz \rightarrow 3, \quad yz \rightarrow 4, \quad xz \rightarrow 5, \quad xy \rightarrow 6. \quad (31)$$

In this notation the fourth-rank tensors can be represented

in the form of 6×6 matrices. Thus, in the most general case only 36 independent components can exist. Actually, the number of independent components is much smaller, since the coupling tensors can be invariant with respect to symmetry-group transformations of the muonium surrounding. The independent components for various symmetry groups are listed in many books (see, e.g., Refs. 1 and 2).

In diamond-structure crystals the Mu-surrounding symmetry in a tetrahedral void is given by the point group T_d . The coupling tensors are given by three constants only, for example:

$$\begin{aligned} Z_{11} = Z_{22} = Z_{33} = Z_{\parallel}, \quad Z_{12} = Z_{21} = Z_{13} = Z_{31} \\ = Z_{23} = Z_{32} = Z_{\perp}, \quad Z_{44} = Z_{55} = Z_{66} = Z_s. \end{aligned}$$

The rest are equal to zero.

Evidently, the specific form of the Hamiltonian (27) for Mu will depend on the acoustic-wave propagation direction and polarization. For isotropic Mu in diamond-structure crystals four independent experiments can be suggested.

1. A longitudinal wave, $\mathbf{k} \parallel z$ ($z \parallel \mathbf{B}$). In this case the Hamiltonian (27) has the form

$$V = \hbar \delta \Omega_{\perp} \sigma_x^{\mu} + \hbar (\delta \Omega_{\parallel} - \delta \Omega_{\perp}) \sigma_z^{\mu} \sigma_z^{\mu} + \mu_0 B \delta g_{\parallel} \sigma_z^{\mu}. \quad (32)$$

2. A longitudinal wave, $\mathbf{k} \perp z$. Let $\mathbf{k} \parallel x$, then

$$V = \hbar \delta \Omega_{\perp} \sigma_x^{\mu} + \hbar (\delta \Omega_{\parallel} - \delta \Omega_{\perp}) \sigma_x^{\mu} \sigma_x^{\mu} + \mu_0 B \delta g_{\perp} \sigma_z^{\mu}. \quad (33)$$

3. A transverse wave, $\mathbf{k} \parallel z$, the polarization $\mathbf{e} \parallel y$. In this case

$$V = \hbar \delta \Omega_s (\sigma_x^{\mu} \sigma_y^{\mu} + \sigma_y^{\mu} \sigma_x^{\mu}) + \mu_0 B \delta g_s \sigma_y^{\mu}. \quad (34)$$

4. A transverse wave, $\mathbf{k} \perp z$, $\mathbf{e} \perp z$. Let $\mathbf{k} \parallel x$, $\mathbf{e} \parallel y$, then

$$V = \hbar \delta \Omega_s (\sigma_x^{\mu} \sigma_y^{\mu} + \sigma_y^{\mu} \sigma_x^{\mu}). \quad (35)$$

We show now how the interaction constants are found. As is well-known, the energy levels of isotropic muonium are (Fig. 2)

$$\epsilon_{1,3} = \hbar \omega_0 / 4 \pm \mu_0 B (1 - \zeta), \quad \epsilon_{2,4} = -(\hbar \omega_0 / 4) \{1 \mp 2(1 + x^2)^{1/2}\}, \quad (36)$$

where ω_0 is the hyperfine splitting frequency, $\zeta = \mu_{\mu} / \mu_0$, and $x = 2\mu_0 B (1 + \zeta) / \hbar \omega_0$. The state vectors, respectively, have the following form

$$\begin{aligned} |1\rangle &= |+\rangle |+\rangle, \quad |2\rangle = 2^{-1/2} (a_+ |+\rangle |-\rangle + a_- |-\rangle |+\rangle), \\ |3\rangle &= |-\rangle |-\rangle, \quad |4\rangle = 2^{-1/2} (a_+ |-\rangle |+\rangle - a_- |+\rangle |-\rangle), \end{aligned} \quad (37)$$

where $\alpha_{\pm} = [1 \pm x / (1 + x^2)^{1/2}]^{1/2}$ and the state vectors are expressed through the products $|\sigma_x^{\mu}\rangle |\sigma_z^{\mu}\rangle$. In what follows we need the expressions for the matrix elements of the operator σ_z^{μ} in the basis (37), therefore we write the matrix in full:

$$\sigma_z^{\mu} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -x/(1+x^2)^{1/2} & 0 & 1/(1+x^2)^{1/2} \\ 0 & 0 & -1 & 0 \\ 0 & 1/(1+x^2)^{1/2} & 0 & x/(1+x^2)^{1/2} \end{pmatrix}. \quad (38)$$

As a rule, for isotropic Mu the frequency $\omega_0 \sim 10^9 - 10^{10} \text{ s}^{-1}$ (see, e.g., Ref. 8). The Larmor frequency of the electron precession in the field $B \sim 100 \text{ G}$ is of order $\omega \sim 10^9 \text{ s}^{-1}$.

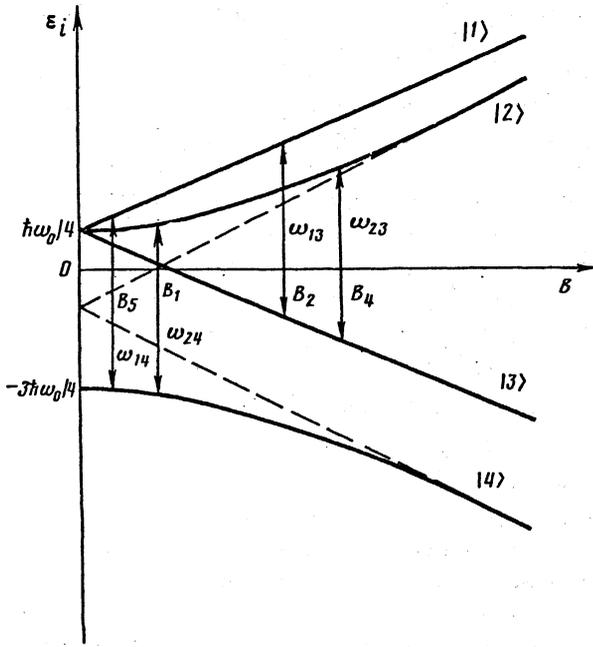


FIG. 2. Muonium hyperfine structure. Arrows indicate possible transitions for $\omega > \omega_0$.

Thus, the μ ASR-experiments should be carried out in the fields of order $B \sim 100$ G.

The longitudinal polarization in isotropic muonium is

$$P_{\parallel}^{(0)}(t) = \frac{1}{2} P_0 \left[1 + \frac{1}{1+x^2} (x^2 + \cos \omega_{24} t) \right]. \quad (39)$$

If the frequency ω_{24} is not resolved in the experiment, the observed polarization is

$$\overline{P_{\parallel}^{(0)}} = \frac{1}{2} P_0 \frac{1+2x^2}{1+x^2}. \quad (40)$$

Consider now the four experiments suggested above.

1. For the Hamiltonian (32) only one nondiagonal matrix element, $\langle 2|V|4 \rangle$, is nonzero; therefore

$$|A| = \langle 2|V|4 \rangle = (2\hbar\delta\Omega_{\perp} x - \delta g_{\parallel} \mu_0 B) / (1+x^2)^{1/2}. \quad (41)$$

Since the audio-signal-generator frequency ω is usually given, the resonance is achieved in a field

$$B_1 = \frac{\hbar\omega_0}{2\mu_0(1+\zeta)} \left[\left(\frac{\omega}{\omega_0} \right)^2 - 1 \right]^{1/2} \approx B_0 \left[\left(\frac{\omega}{\omega_0} \right)^2 - 1 \right]^{1/2}, \quad (42)$$

where B_0 is the hyperfine field at the muon. We disregard hereafter the small quantity ζ in comparison with unity.

From Eq. (38) we have $\sigma_{44} = -\sigma_{22} = x/(1+x^2)^{1/2}$ and $\sigma_{24} = 1/(1+x^2)^{1/2}$. Thus the resonance correction δP_{\parallel} and the minimum depth in $\langle P_{\parallel} \rangle$ are fully determined.

2. In this case the Hamiltonian (33) has two nonzero nondiagonal elements

$$\langle 1|V|3 \rangle = \hbar(\delta\Omega_{\parallel} - \delta\Omega_{\perp}), \quad (43)$$

$$\langle 2|V|4 \rangle = [\hbar(\delta\Omega_{\parallel} - \delta\Omega_{\perp})x - \delta g_{\perp} \mu_0 B] / (1+x^2)^{1/2}. \quad (44)$$

It is the transition $2 \leftrightarrow 4$ that is at resonance with the acoustic wave in the field (42) is $2 \leftrightarrow 4$ so that the matrix element (44) is determined. The transition $1 \leftrightarrow 3$ is at resonance in the field

$$B_2 \approx \hbar\omega / 2\mu_0. \quad (45)$$

In this case $\sigma_{11} = -\sigma_{33} = 1$ and $\sigma_{13} = 0$.

3. The "richest" experiment. Four matrix elements of the Hamiltonian (34) are nonzero:

$$\langle 1|V|2 \rangle = -(i/2^{\hbar}) [a_+ \hbar\delta\Omega_3 + a_- (\hbar\delta\Omega_3 + \delta g_3 2\mu_0 B)], \quad (46)$$

$$\langle 2|V|3 \rangle = (i/2^{\hbar}) [a_- \hbar\delta\Omega_3 + a_+ (\hbar\delta\Omega_3 - \delta g_3 2\mu_0 B)], \quad (47)$$

$$\langle 1|V|4 \rangle = (i/2^{\hbar}) [a_- \hbar\delta\Omega_3 - a_+ (\hbar\delta\Omega_3 + \delta g_3 2\mu_0 B)], \quad (48)$$

$$\langle 3|V|4 \rangle = -(i/2^{\hbar}) [a_+ \hbar\delta\Omega_3 - a_- (\hbar\delta\Omega_3 - \delta g_3 2\mu_0 B)]. \quad (49)$$

Correspondingly, for the transition $1 \leftrightarrow 2$ Eq. (38) yields $\sigma_{12} = 0$, and the resonance is achieved in the field

$$B_3 \approx (\hbar\omega / \mu_0) (1 - \omega / \omega_0) (1 - 2\omega / \omega_0)^{-1}. \quad (50)$$

For the transition $2 \leftrightarrow 3$ we also have $\sigma_{23} = 0$ and the resonance in the field

$$B_4 = (\hbar\omega / \mu_0) (1 + \omega / \omega_0) (1 + 2\omega / \omega_0)^{-1}. \quad (51)$$

The transitions $1 \leftrightarrow 2$ and $2 \leftrightarrow 3$ are inside the triplet. For the triplet-singlet transitions we find that the transition $1 \leftrightarrow 4$ is at resonance in the field

$$B_5 = (\hbar\omega / \mu_0) (1 - \omega / \omega_0) (1 - 2\omega / \omega_0)^{-1}, \quad (52)$$

and the transition $3 \leftrightarrow 4$ in the field

$$B_6 = (\hbar\omega / \mu_0) (1 - \omega / \omega_0) (2\omega / \omega_0 - 1)^{-1}. \quad (53)$$

The singularities in Eqs. (50), (52) and (53) at $\omega = \omega_0/2$ are due to neglect of ζ . As will be shown below, this does not lead to any misunderstandings.

4. In this experiment the Hamiltonian (35) has one off-diagonal matrix element

$$\langle 1|V|3 \rangle = i\hbar 2\delta\Omega_3. \quad (54)$$

Thus, in the field B_2 given by Eq. (45) the constant $\delta\Omega_3$ is found in a straightforward manner.

Evidently, at a given frequency ω of the audio-signal generator we cannot excite all transitions. Consequently, the information obtained in experiment will not be excessive.

First of all, note that if $\omega < \omega_0$ the acoustic resonance can be observed only for intratriplet transitions. There are three of them: $1 \leftrightarrow 2$, $2 \leftrightarrow 3$, and $1 \leftrightarrow 3$. As is seen, four experiments are possible: the experiment 4 [the transition $1 \leftrightarrow 3$, Eq. (54)] allows us to find the constant $\delta\Omega_3$. It is usually assumed that the material-tensor traces over the first two indices equal zero,¹ i.e. $z_{ilm} = 0$ and $F_{ilm} = 0$. Then, knowing $\delta\Omega_3$, we find $\delta\Omega_{\parallel} = -\delta\Omega_3$.

In the experiment 3, by exciting the transition $1 \leftrightarrow 2$ or $2 \leftrightarrow 3$, we find δg_3 and, correspondingly, $\delta g_{\parallel} = -\delta g_3$. In the remaining experiment 2 [the transition $1 \leftrightarrow 3$, Eq. (43)] the constant $\delta\Omega_1$ is determined. Thus, by exciting the transitions inside the triplet we cannot find the constant δg_{\perp} . However, these experiments are convenient in that they allow to use low-frequency audio-signal generators. Note that at a generator frequency $\omega \ll \omega_0$ we get in the experiment 3, as seen from Eqs. (50) and (51), $B_3 \approx B_4 \approx 2B_2$, i.e. two transitions are at resonance simultaneously, therefore the observed picture is more complicated than that given by Eqs. (22) and (25). This is to be born in mind in setting the experiment: the ratio ω/ω_0 should not be smaller than the minimum width given by Eq. (25).

At high frequencies of the ultrasound generator two frequency ranges can be singled out when different transitions

are excited at a given frequency ω .

As seen from (49), resonance at the transition $3 \leftrightarrow 4$ is possible if $\frac{1}{2} < \omega/\omega_0 < 1$. In this case, alongside with the transition $3 \leftrightarrow 4$, it is possible to excite also the transitions $1 \leftrightarrow 3$ and $2 \leftrightarrow 3$. Let, for example, $\omega/\omega_0 = 0.6$, then the transition $3 \leftrightarrow 4$ is at resonance in the field $B_6 = 2\hbar\omega/\mu_0 = 4B_2$, and the transition $2 \leftrightarrow 3$ in the field $B_4 \approx 1.45B_2$. For normal muonium (Mu) in silicon we have $\omega_0 \approx 1.26 \cdot 10^{10} \text{ s}^{-1}$ ($\nu_0 = 2006 \text{ MHz}$),⁸ so that $\omega = 0.6\omega_0 = 7.56 \cdot 10^9 \text{ s}^{-1}$ (or $\nu = 1.2 \text{ GHz}$). The corresponding resonance field is $B_2 \approx 409 \text{ G}$. As we see, both the audio-signal generator frequency and magnetic fields are quite achievable. Unfortunately, as in the case of low frequencies (intratriplet transitions), the parameter δg_{\perp} remains undetermined.

Full information is obtained only in the case when $\omega > \omega_0$. In fact, in the experiment 4 the transition $1 \leftrightarrow 3$ is excited in the field B_2 given by (45), and the constant $\delta\Omega_3$ is found ($\delta\Omega_{\parallel} = -\delta\Omega_3$). Next, in the experiment 2, exciting again the transition $1 \leftrightarrow 3$, we find the constant $\delta\Omega_1$ [see Eq. (43)]. If, in the same experiment, we excite the transition $2 \leftrightarrow 4$ in field $B_1 < B_4$ [see (42)], we find the parameter δg_{\perp} . In the experiment 1 the transition $2 \leftrightarrow 4$ is excited in the field B_1 and the constant δg_{\parallel} is found (correspondingly, $\delta g_3 = -\delta g_{\parallel}$). Furthermore, extra checking measurements of the constants δg_3 and $\delta\Omega_3$ can be conducted in experiment 3. In fact, the transition $2 \leftrightarrow 3$ is excited in field $B_4 > B_2$ and the transition $1 \leftrightarrow 4$ in field $B_5 < B_2$. These two measurements determine the constants $\delta\Omega_3$ and δg_3 [see Eqs. (47) and (48)].

The frequency and field values for the Mu atom in Si are roughly as follows. Let, for example, ω be $1.5 \cdot 10^{10} \text{ s}^{-1}$ ($\nu = 2.4 \text{ GHz}$), then the resonant field values are $B_1 \approx 430 \text{ G}$ (the transition $2 \leftrightarrow 4$), $B_2 \approx 811 \text{ G}$ (the transition $1 \leftrightarrow 3$), $B_4 \approx 1050 \text{ G}$ (the transition $2 \leftrightarrow 3$), and $B_5 \approx 223 \text{ G}$ (the transition $1 \leftrightarrow 4$).

Thus, μASR -experiments allow not only to find new constants of the spin-phonon interaction of the Mu atom in crystals for μSR -experiments, but also to conduct precision

measurements of already known parameters of the Mu-atom hyperfine structure. In particular, by exciting the transition $1 \leftrightarrow 3$, we find the muonium electron g -factor. The spin-phonon interaction constants are not only of purely academic interest, they are important from the point of view of understanding the behavior of light impurities in semiconductors and dielectrics. In fact, the spin-phonon interaction constants depend both on the muon localization site and its state in the lattice. In particular, the μASR -experiments will probably clarify the nature of normal muonium (Mu) in diamond- and zinc-blende-type crystals. It is also evident that the μASR -experiments with anisotropic Mu^* give more information. The general formulas (22) and (25) are valid, of course, but the specific formulas for the spin-phonon interaction Hamiltonian and its matrix elements are more complicated. Though it is not difficult to find them, we have omitted these calculations for the sake of simplicity and clarity. Note that the spin-phonon interaction constants of the Mu and Mu^* atoms in Si, Ge, and diamond have been measured indirectly using the temperature dependence of the hyperfine interaction constants (see, e.g., Ref. 8). Attempts have been made to study the acoustic spin resonance in the hyperfine structure of the Mu atom in quartz.⁹

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