

# Theory of the interaction of atoms (muonium, positronium) with magnetized substances

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An analysis is made of the interaction of muonium and positronium atoms with magnetized substances. It is shown that the effective field generated in such substances as a result of the exchange scattering gives rise to the linear Zeeman effect in the positronium levels with  $m = \pm 1$ . Equations are derived for the evolution of the polarization characteristics of muonium. It is shown that the effective exchange field alters considerably the precession frequency of a  $\mu^+$  meson.

1. One of the present authors<sup>[1,2]</sup> has shown that a beam of electrons (including electrons in atoms) traversing a magnetized substance (gas, liquid, or solid) experiences not only the usual macroscopic magnetic field  $B$  but also an additional quasimagnetic effective field  $G$ . The latter field appears because of the exchange Coulomb scattering of electrons. This field resembles the molecular Weiss field responsible for ferromagnetism. More recently, Lambert<sup>[3]</sup> reached the same conclusion in related to optically pumped gases.

The interest in the interaction between atoms and substances containing polarized electrons is increasing. For example, D'yakonov and Perel<sup>[4]</sup> analyzed the interaction between impurity centers and an electron gas in a semiconductor polarized by optical pumping. Ivanter<sup>[5]</sup> considered the interaction of muonium with magnetized substances. However, the exchange field was ignored in these investigations.

In the present paper we shall use the example of the interaction of muonium and positronium with substances containing polarized electrons to derive the equations of motion for the spin density matrix of atoms. We shall show that allowance for the effective field  $G$  alters considerably the frequency of precession of a  $\mu^+$  meson compared with the results obtained in<sup>[5]</sup>. We shall also demonstrate that, in contrast to the usual field  $B$ , the field  $G$  gives rise to the linear Zeeman effect in positronium.

2. Let us assume that an atom of, say, muonium or positronium interacts with a magnetized substance, i.e., with a medium containing polarized electrons. We shall first consider the interaction of atoms with a polarized gas. In this case, we can use the highly developed density matrix theory which describes the shifts and broadening of the atomic levels because of collisions in a polarized medium.<sup>[2,6]</sup> According to this theory, the spin density matrix of an atom in an  $s$  state is described by

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}(H\rho - \rho H^*) + \left(\frac{d\rho}{dt}\right)_c, \quad (1)$$

where  $H = (1/4)\hbar\omega_0\sigma_1\sigma_2 - \mu_1\sigma_1B - \mu_2\sigma_2B + (1/2)i\Gamma$ ,  $\omega_0$  is the hyperfine splitting frequency,  $\mu_1$  is the magnetic moment of an electron,  $\mu_2$  is the magnetic moment of a  $\mu^+$  meson (or of a positron or a nucleus with spin  $1/2$ ),  $\Gamma$  is the decaying part of the Hamiltonian. If, for example, in the case of positronium  $|\varphi_n\rangle$  are the eigenfunctions of the operator  $(1/4)\hbar\omega_0\sigma_1\sigma_2$ , it follows that  $\Gamma|\varphi_n\rangle = \Gamma_n|\varphi_n\rangle_1$ , where  $\Gamma_n$  is the decay width of triplet or singlet states. The term  $(d\rho/dt)_c$  describes the collision-induced change in the density matrix of an atom.

The expression for  $(d\rho/dt)_c$  was first derived for a polarized gas in<sup>[6]</sup> from an analysis of spin-exchange collisions of two hydrogen-like atoms. Using the results of this analysis, we can write the collision term  $(d\rho/dt)_c$  in the following form (for the sake of simplicity we shall assume that the atoms in the medium have one unpaired electron each and there is no spin-orbit scattering):

$$\left(\frac{d\rho}{dt}\right)_c = \frac{i\alpha}{2}[\sigma_1P, \rho] + \frac{1}{4T_1}\{-3\rho + \sigma_{1m}\rho\sigma_{1m} + \sigma_1P\rho + \rho\sigma_1P - i\epsilon_{nst}\sigma_{1n}\rho\sigma_{1n}P\}, \quad (2)$$

where  $\epsilon_{nst}$  is a totally antisymmetric unit tensor,  $P$  is the polarization vector of the electrons in the medium,  $T_1$  is the spin-exchange relaxation time,

$$1/T_1 = \langle vN\sigma_{SF} \rangle, \quad (3)$$

$\langle \rangle$  represents averaging over the relative velocities  $v$  of the motion of a given atom (mesic atom) and atoms (electrons) in the medium,  $N$  is the density of atoms in the medium,  $\sigma_{SF}$  is the cross section for the exchange scattering accompanied by spin flip:

$$\sigma_{SF} = \frac{1}{4} \int d\Omega |f_3 - f_1|^2 = \frac{\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \sin^2(\delta_l^3 - \delta_l^1), \quad (4)$$

$f_3$  and  $\delta_l^3$  are the amplitude and phase of the triplet scattering of an atom (mesic atom) by an atom (electron) in a gas,  $f_1$  and  $\delta_l^1$  are the corresponding parameters of the singlet scattering,  $\alpha$  is the spin-exchange interaction constant:

$$\alpha = \left\langle \frac{vN\pi}{2k^2} \sum_{l=0}^{\infty} (2l+1) \sin 2(\delta_l^3 - \delta_l^1) \right\rangle. \quad (5)$$

The processes resulting in the loss of an atom (mesic atom) as a result of detachment of an electron or a chemical reaction, which are independent of the spin state of the colliding atoms, may be included in the definition of the decay width  $\Gamma$  and, therefore, these processes are ignored in the expression for  $(d\rho/dt)_c$ . However, if inelastic processes depend on the spin state of the colliding particles, the phases  $\delta$  become complex. For the sake of simplicity we shall exclude this case from our consideration.

Substituting Eq. (2) into Eq. (1), we obtain the following equation for the spin density matrix of an atom (muonium, positronium)

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}(H_{\text{eff}}\rho - \rho H_{\text{eff}}^*) + \frac{1}{4T_1}\{-3\rho + \sigma_{1m}\rho\sigma_{1m} + \sigma_1P\rho + \rho\sigma_1P - i\epsilon_{nst}\sigma_{1n}\rho\sigma_{1n}P\}, \quad (6)$$

where  $H_{\text{eff}}$  denotes the effective Hamiltonian of an atom in a medium defined by

$$H_{\text{eff}} = \frac{1}{4}\hbar\omega_0\sigma_1\sigma_2 - \mu_1\sigma_1B - \frac{1}{2}\hbar\alpha\sigma_1P - \mu_2\sigma_2B - \frac{1}{2}i\Gamma. \quad (7)$$

According to Eqs. (6) and (7), an electron of an atom (mesic atom) is acted upon not only by the magnetic field  $B$  but also by an additional field proportional to the degree of polarization of electrons in the medium and governed by the amplitudes of the Coulomb exchange interaction. It should be noted that although Eqs. (2)–(7) have been derived for a gas, they may be applied to the interaction of an atom with a polarized gas of electrons in a semiconductor if the scattering amplitudes are understood to be the amplitudes of the scattering of an atom by electrons in the medium. In the general case of an arbitrary medium, the exchange field still acts on atomic electrons but the actual expression for the constant  $\alpha$  governing the exchange field is generally unknown (for example, in the case of an atom at rest in a medium, this constant is governed by the overlap integral, which may be compared with the case of ferromagnetism).<sup>[7]</sup>

3. We shall now consider the action of the exchange field on positronium. It is well known<sup>[8,9]</sup> that because the signs are opposite and the absolute values of the magnetic moments of an electron and a positron are equal, the positronium states with magnetic quantum numbers  $m = +1$  and  $m = -1$  are not split in the magnetic field  $B$ . However, we can easily show that such splitting may be caused by the exchange field. For the sake of simplicity, we shall assume that the real part of the scattering amplitude is much greater than its imaginary part so that  $\alpha \gg 1/T_1$  (an estimate of the order of magnitude of the spin-exchange interaction constant  $\alpha$  and of the spin-exchange relaxation time  $T_1$  will be given later). Therefore, ignoring in Eq. (6) the terms proportional to  $1/T_1$ , we obtain the following equation for the density matrix of an atom

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}(H_{\text{eff}}\rho - \rho H_{\text{eff}}^{\dagger}), \quad (8)$$

i.e., in the case under consideration we can describe the action of a medium on positronium by the effective Hamiltonian  $H_{\text{eff}}$ . If we select the quantization axis along the field  $B$  and if we assume that the vector of the steady-state polarization of the medium is also directed along  $B$ , we find that  $H_{\text{eff}}$  becomes

$$H_{\text{eff}} = \frac{1}{2} \hbar \omega_0 \sigma_1 \sigma_2 - \mu_{1\text{eff}}(B) \sigma_{1z} B - \mu_2 \sigma_{2z} B - \frac{1}{2} i \Gamma, \quad (9)$$

where the effective moment of an electron in the medium is

$$\mu_{1\text{eff}}(B) = \mu_1 + \hbar \alpha P / 2B. \quad (10)$$

We can see that because of the exchange interaction the effective magnetic moment of an electron belonging to an atom is not equal to its vacuum value  $\mu_1$ . Since  $|\mu_{1\text{eff}}| \neq |\mu_2|$ , the splitting is now allowed. Using the well-known solution of the problem of behavior of a system of two spins of different moments bound by the hyperfine interaction and subjected to an external magnetic field,<sup>[10,11]</sup> we obtain the following expression for the positronium levels:

$$E_{m=\pm 1} = \frac{\hbar \omega_0}{4} \mp \frac{\hbar \alpha}{2} P. \quad (11)$$

The difference between these levels is

$$E_{m=+1} - E_{m=-1} = -\hbar \alpha P \quad (12)$$

and it is governed solely by the exchange interaction. The presence of a nonzero difference between the levels  $E_{m=\pm 1}$  means that a study of a reduction in the contribution of  $3\gamma$  collisions to the spectrum of annihilation quanta considered as a function of the frequency of an alternating field inducing the transitions ( $m = \pm 1$ )

→ ( $m = 0$ ) should reveal two maxima separated by an interval equal to the difference between the levels  $E_{m=\pm 1}$ . In order to estimate the order of magnitude of the splitting, we shall use the data on the conversion cross section of positronium in oxygen. According to the results reported in<sup>[9,12]</sup>, this cross section is dominated by the collisions involving electron exchange and its value is  $\sigma \approx 3.6 \times 10^{-19} \text{ cm}^2$ . If we assume that the main contribution to the scattering is made by the s waves (for positronium the velocity is  $v \approx 6.6 \times 10^6 \text{ cm/sec}$  and the wavelength is  $\lambda = 10^{-6} \text{ cm}$ , which is much greater than the dimensions of gas atoms), we find from Eqs. (4) and (5) that the order of magnitude of the exchange constant is

$$\alpha \approx \hbar \sqrt{\sigma} N / m \approx 10^{-10} N \quad (13)$$

and that  $\alpha \approx 10^2 T_1^{-1}$ , i.e., the condition  $\alpha \gg 1/T_1$  is satisfied. For a gas with  $N = 10^{18} - 10^{19} \text{ cm}^{-3}$  the frequency difference of interest to us is

$$\Delta \omega = |E_{m=+1} - E_{m=-1}| / \hbar = \alpha P \approx (10^8 - 10^9) P$$

and it depends strongly on the degree of polarization of electrons in a gas (or a condensed substance). If  $P = 1$ , we find that  $\Delta \omega = 10^8 - 10^9 \text{ sec}^{-1}$ .

4. We shall now consider the equations which describe the evolution of the polarization characteristics of muonium in a magnetized substance. In order to do this, we shall multiply Eq. (6) by  $\sigma_1$  and calculate the trace. In this way, we obtain

$$\frac{dP_1}{dt} = -\frac{\omega_0}{2} \text{Tr}[\sigma_1 \times \sigma_2] \rho - \frac{2\mu_1}{\hbar} [B \times P_1] - \alpha [P \times P_1] - \frac{P_1 - P}{T_1} - \frac{P_1}{\tau}, \quad (14)$$

where  $P_1 = \text{Tr} \sigma_1 \rho$  is the polarization vector of a muonium electron and  $\tau$  is the muonium lifetime.

Multiplying Eq. (6) by  $\sigma_2$ , we obtain an equation which describes the evolution of the polarization vector  $P_2$  of a  $\mu^+$  meson:

$$\frac{dP_2}{dt} = \frac{\omega_0}{2} \text{Tr}[\sigma_1 \sigma_2] \rho - \frac{2\mu_2}{\hbar} [B P_2] - \frac{P_2}{\tau}. \quad (15)^*$$

Multiplying finally Eq. (6) by  $\sigma_{1m} \sigma_{2\lambda}$ , we obtain the following equation for the correlation tensor

$$Q_{m\lambda} = \text{Tr} \sigma_{1m} \sigma_{2\lambda} \rho:$$

$$\frac{dQ_{m\lambda}}{dt} = \frac{\omega_0}{2} \epsilon_{m\lambda n} (P_1 - P_2)_n - \frac{2\mu_1}{\hbar} \epsilon_{min} B_i Q_{n\lambda} - \alpha \epsilon_{min} P_i Q_{n\lambda} - \frac{2\mu_2}{\hbar} \epsilon_{in\lambda} B_i Q_{m\lambda} - \left( \frac{1}{T_1} + \frac{1}{\tau} \right) Q_{m\lambda} + \frac{1}{T_1} P_m P_{2\lambda}, \quad (16)$$

where  $\epsilon_{m\lambda n}$  is a totally antisymmetric unit tensor.

Comparing Eqs. (14) and (16) with the phenomenological equations (5) and (6) in Ivanter's paper,<sup>[5]</sup> we find that our Eqs. (14) and (16) differ by the presence of terms proportional to the exchange constant  $\alpha$  and describing the action of the exchange field on the electron spin. (The physical origin of this field is the fact that in the exchange scattering the change from the direction of the spin of an atomic electron to the direction of the spin of an electron in the medium is accompanied by the rotation of the spin of the atomic electron about the polarization vector of the electron in the medium. The coherent addition of these elementary rotations results in the precession of the spin of the atomic electron in the effective field governed by the exchange interaction.) It also follows from Eqs. (14) and (16) that in the case of interactions of muonium with a gas (including a gas electrons in a semiconductor) and spin relaxation solely because of the spin exchange (no spin-orbit scattering),

the phenomenological constants  $\nu$  and  $f$  introduced in<sup>[5]</sup> are equal:  $\nu = f = (1/2)T_1$ .

If the magnetic field  $B$  and the polarization vector of electrons in the medium  $P$  are oriented along the same direction, we find that—as mentioned in Sec. 3 and as follows directly from Eqs. (14) and (16)—the action of the exchange field can be regarded as a change in the magnetic moment of an electron in the medium. In this case, Eqs. (14) and (16) generalized to arbitrary media can be written in the form

$$\frac{dP_i}{dt} = -\frac{\omega_0}{2} \text{Tr}[\sigma_i \sigma_2] \rho - \frac{2\mu_{1\text{eff}}}{\hbar} [BP_i] - 2\nu \left[ P_i - \frac{f}{\nu} P \right] - \frac{P_i}{\tau}, \quad (17)$$

$$\frac{dQ_{m\lambda}}{dt} = \frac{\omega_0}{2} \epsilon_{m\lambda n} (P_1 - P_2)_n - \frac{2\mu_{1\text{eff}}}{\hbar} \epsilon_{m1n} B_i Q_{n\lambda} - \frac{2\mu_2}{\hbar} \epsilon_{n12} B_i Q_{m\lambda} - 2\nu \left[ Q_{m\lambda} - \frac{f}{\nu} P_m P_{2\lambda} \right] - \frac{1}{\tau} Q_{m\lambda}. \quad (18)$$

Since these expressions are identical with the equations discussed in<sup>[5]</sup>, their solutions are given by Eqs. (13)–(19) in<sup>[5]</sup> (we must bear in mind that the magnetic moment of an electron is now  $\mu_{1\text{eff}}$ ); it is sufficient to redefine the constants  $\xi$  and  $\omega'$  in<sup>[5]</sup>, which now become

$$\xi = -\mu_2/\mu_{1\text{eff}}, \quad \omega_m' = -2\mu_{1\text{eff}} B_m/\hbar. \quad (19)$$

It should be noted (Sec. 3) that the exchange constant  $\alpha$  is generally much greater than the relaxation constants.<sup>1)</sup> In this case, muonium in a polarized medium can be described by an effective Hamiltonian of the type given by Eq. (9) and the energy spectrum of this Hamiltonian is

$$E_{1, \pm 1} = \frac{1}{2} \hbar \omega_0 \mp (\mu_{1\text{eff}} + \mu_2) B, \\ E_{1(0), 0} = \frac{\hbar \omega_0}{4} \left\{ -1 \pm 2 \left[ 1 + \frac{4(\mu_{1\text{eff}} - \mu_2)^2 B^2}{\hbar^2 \omega_0^2} \right]^{1/2} \right\}, \quad (20)$$

where the subscript  $F$  in  $E_F$ ,  $m_F$  is the total moment of muonium and  $m_F$  is its projection along the quantization axis; the constant  $\Gamma$  describing the width of the levels is omitted (for the sake of simplicity).

The positions of the energy levels of muonium in a medium depend on the exchange field [Eq. (20)]. Since the difference between the energies of the levels gives rise to precession frequencies of the spin of a  $\mu^+$  meson,<sup>[13]</sup> the exchange field can be studied not only by measuring the splitting of the muonium levels by resonance methods but also by investigating the two-frequency precession of this meson.<sup>[13]</sup> By way of example, we shall consider the case when the splitting of the muonium levels in fields is much less than the hyperfine splitting. Applying Eq. (20), we can use the two-frequency precession approach<sup>[13]</sup>

$$\omega = \frac{E_{1,1} - E_{1,-1}}{2\hbar}, \quad \Omega = \frac{2E_{1,0} - (E_{1,1} + E_{1,-1})}{2\hbar}$$

to obtain the following expressions for the precession frequencies of a  $\mu^+$  meson:

$$\omega = -\frac{(\mu_{1\text{eff}} + \mu_2) B}{\hbar}, \quad \Omega = \frac{(\mu_{1\text{eff}} - \mu_2)^2 B^2}{\hbar^2 \omega_0}; \quad (21)$$

it is worth recalling that  $\mu_{1\text{eff}} = \mu_1 + \hbar \alpha P/2B$ .

According to Eq. (21), we can select such a value of

the field  $B$  in which  $\mu_{1\text{eff}} = -\mu_2$  and the frequency  $\omega$  vanishes. In this case, the precession of a  $\mu^+$  meson has a very low frequency:  $\Omega = 4\mu_2^2 B^2/\hbar^2 \omega_0$ . In the opposite case of strong fields, we obtain

$$E_{1, \pm 1} = \frac{\hbar \omega_0}{4} \mp \left( \mu_1 B + \frac{\hbar \alpha}{2} P + \mu_2 B \right), \\ E_{1(0), 0} = -\frac{\hbar \omega_0}{4} \pm \left( \mu_1 B + \frac{\hbar \alpha}{2} P - \mu_2 B \right). \quad (22)$$

These formulas also apply in the case when only the field  $B$  is high and the exchange field is arbitrary. It follows from Eq. (22) that the shift of the energy levels and, consequently, of the precession frequencies of a  $\mu^+$  meson is proportional to the exchange constant  $\alpha$  and to the degree of polarization  $P$ , and in the latter case it does not contain the factor  $\omega_0$  [compare with Eq. (20) in Ivanter's paper<sup>[5]</sup>].

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$$*[\sigma_1 \sigma_2] \equiv \sigma_1 \times \sigma_2.$$

<sup>1)</sup>Near the particular value of the phase difference  $\delta_0^3 - \delta_0^1 = \pi/2$  [see Eqs. (3)–(5)] in the case of s-type scattering the exchange constant  $\alpha$  may be also much smaller than  $1/T_1$ .

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