Charge exchange of muons in gases. Kinetic equations

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(Received 3 June 1983)

Kinetic equations for the spin-density operators of the diamagnetic and paramagnetic states of the positive muon are obtained for the description of the slowing-down process encountered when high-energy muons thermalize in a single-component gas. The motion of this two-species system is generated by the Liouville superoperators associated with the diamagnetic and paramagnetic spin Hamiltonians and by time-dependent rate superoperators which depict the probabilities per collision that an electron is captured or lost. These rates are translational averages of the appropriate Boltzmann collision operators. That is, they are momentum and position integrals of the product of either the electron capture or loss total cross section with the single-particle translational density operators for the muon (or muonium) and a gas particle. These rates are time dependent because the muon (or muonium) translational density operator is time dependent. The initial amplitudes and phases of the observed thermal spin polarization in muon-spin-rotation (μSR) experiments are then obtained in terms of the spin-density operators emerging from the stopping regime.

I. INTRODUCTION

In the typical muon-spin-rotation (μSR) experiment[1] the time dependence of the spin polarization of an ensemble of muons is followed by observing the ensemble of decay positrons emitted along the direction of the muon spin vectors. Two muon spin states are experimentally resolved,[1] that is, a diamagnetic state and a paramagnetic state. This resolution is accomplished by observing the characteristic Larmor frequencies associated with each state. Under current experimental conditions[1] the chemical identity of the diamagnetic spin state is not resolved, that is, it may be the bare muon or it may be a chemical compound in which the muon is in a diamagnetic environment. On the other hand, the chemical identity of the paramagnetic state is known, that is, it is the electronic ground state of the muonium atom (positive muon equivalent of the hydrogen atom). Generally, the time resolution[1] of the experiments is on the order of nanoseconds which precludes direct studies of the stopping region of the highly energetic incoming muon since, in gases, the time scale[2–4] of this slowing down process is also of the order of nanoseconds. Indeed, in condensed matter the time scale[5,6] is of the order of picoseconds. What is observed then is the time dependence of the thermalized products of this stopping regime. That is, the outcome of this slowing-down region acts as the initial condition of the experiment. Thus, detailed knowledge of this region can only be inferred from experiments. In particular, the amplitudes and phases of the various signals at the end of the thermalization process can be obtained. It is therefore of considerable interest to have a theoretical basis for understanding the effects that the thermalization of the translational motion has upon the spin dynamics of the muon.

Stopping processes by which highly energetic particles thermalize in matter have been studied in great detail for many years.[3,7–12] In these treatments the main concern has usually been centered around the kinetic energy loss felt by the incoming particle and its effect on the surrounding media. Little attention has been accorded to the effect of the stopping process on the internal states of the incoming particle since such changes are usually not measured. An exception to this generalization is, of course, μSR where the observable change of interest is an internal state of the incoming particle, namely, the muon spin vector. In general, the effect of the stopping process on the muon spin vector is not well known for either the dilute or the condensed phase. It has been the subject of much debate in liquids[5,6] and has been qualitatively understood in gases.[2,4]

This is the first of a series of papers whose objective is to quantitatively describe the effects that the stopping process has on the spin dynamics of the muon. The purpose of this first paper is twofold. That is, first of all, it is to develop the theoretical framework, namely, rate equations based upon the Boltzmann equation, which describes the charge-exchange regime. Secondly, it is to relate the formal solutions of these rate equations to the observed amplitudes and phases of the experimental μSR signal. Explicit formulas for these amplitudes and phases are developed in the second paper[13] of this series based upon a time-independent rate approximation. Such a description, while providing an explanation of the gas phase experiments, may be suggestive of the physics occurring in more condensed matter.

There are four main regions in the stopping process of a highly energetic charged particle in dilute gases. The first region, from time 0 to t_1, is the Bethe-Bloch regime where the dominant energy-loss mechanism consists of collisions which ionize the moderating gas atoms. It is assumed that such ionizing collisions have no effect on the muon spin vector. Thus, the time dependence of the polarization is due solely to the diamagnetic muon Larmor precession about the external magnetic field. Since the magnetic fields of experimental interest are of the order of 100 G then the Larmor frequency is of the order of 10^7/sec [see

Eq. (4.5)]. On the other hand, the time duration[1] of the Bethe-Bloch region is of the order of nanoseconds. Thus, the polarization is essentially a constant in the first region. The second region, from time t_1 to t_2, $t_C = t_2 - t_1$, is the charge-exchange regime where repeated charge-exchange collisions produce an oscillation between the chemical species in which the muon is in a diamagnetic environment and the chemical species, namely, muonium, in which it is paramagnetic. Other processes, namely, elastic, inelastic, and reactive collisions may occur in this region. However, the spin dynamics is dominated by the charge-exchange cycles which inter-relate the spin dynamics of the diamagnetic and paramagnetic states. The time span of this charge-exchange region is also of the order of or less than a nanosecond. Thus, the muonium hyperfine frequency, which is 28×10^9/sec, plays an important role in this region. It is the only frequency of importance. There are two mechanisms that lead to a loss of signal amplitude in this region. One is an ensemble mechanism in which the spin dephasing is caused by the different trajectories experienced by each individual muon. The other mechanism involves a free-flight exchange of polarization between the muon spin and the electron spin dictated by the hyperfine interaction. This leads to a loss of signal amplitude as the electrons carry off whatever polarization they have acquired when muonium loses its electron upon a collision with a gas atom. The third regime, from time t_2 to t_3, is the thermalization region where again elastic, inelastic, and possibly reactive collisions occur. However, in this region there is no feedback of polarization to and from the different chemical species of the muon as no further charge-exchange cycles occur. The spin dynamics of the different chemical species are then governed separately by their respective spin Hamiltonians. Thus, the polarization emerging from the charge-exchange regime acts as the initial conditions for this third region and the subsequent fourth or thermal region.

Since the stopping process of highly energetic charged particles in dilute gases involves a series of binary collisions, then it is appropriate to use the Boltzmann equation[14–16] to describe this thermalization. Starting with the full $(N+1)$-particle density operator for the gas plus muon system, two first-order Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) equations[17] for the diamagnetic muon and paramagnetic muonium species are obtained in Sec. II A. The generalized Boltzmann ansatz[17] is then invoked to produce a pair of coupled generalized Boltzmann equations for these two species. These equations couple the translational motion of the two species to the spin dynamics of the muon and an associated electron. Since it is the polarization of the muon spin vector that is observed experimentally[1] then it is appropriate to average over all translational degrees of freedom. As a result the spin degrees of freedom of the diamagnetic and paramagnetic muon species are coupled through a pair of rate equations. The dynamics are generated by the spin Liouville superoperators associated with the free particles and by time-dependent rate superoperators which describe the probability per collision that an electron is captured or lost. These latter processes arise totally from the translational motion and are explicitly considered in Sec. III. The rate equations are developed using the Schrödinger picture in which the successive collisional aspect of these Boltzmann equations is not explicit. To elucidate this aspect of the stopping process, the rate equations are rewritten in Sec. II B using an interaction picture which explicitly demonstrates the sequence of binary collisions that these dynamical equations describe. Finally, in Sec. IV, the observed thermal experimental signals are related to elements of the density operators which emerge at the end of the stopping process as described in Sec. II. Here, it is assumed that only the electronic ground state of paramagnetic muonium contributes to the initial signal since the hyperfine interaction for electronically excited states is too weak to allow exchange of polarization between the muon and the electron during the time span of the charge-exchange region. After thermalization, it is also assumed that the observed paramagnetic signal arises solely from ground-state muonium.

II. BOLTZMANN EQUATION FOR THE CHARGE-EXCHANGE REGION

A. Schrödinger picture

The total Hamiltonian, H_T, which describes the incoming highly energetic muon and the single-component moderating gas can be written in two forms, namely,

$$H_T = K_\mu + H_\mu^{\mathrm{sp}} + H_G + \sum_{i=1}^{N} V_{i\mu}$$
$$= K_{\mathrm{Mu}} + H_{\mathrm{Mu}}^{\mathrm{sp}} + H_G' + \sum_{i=1}^{N} V_{i\mathrm{Mu}} . \tag{2.1}$$

Here, K_μ is the kinetic energy of the bare muon while H_μ^{sp} is its spin Hamiltonian. Associated with these operators are the Hamiltonians H_G and $V_{i\mu}$ for the moderating gas and for the interaction between the gas atoms and the muon, respectively. The Hamiltonian H_G contains the kinetic energies of the gas atoms as well as all the two-particle potentials which give rise to the thermal gas-gas collisions. Also included in this Hamiltonian are the spin degrees of freedom of the electrons of the gas atoms. The potentials between the gas atoms and the muon are assumed to be translational in nature. They do not affect the spin degrees of freedom of either the muon or the electrons. The second form of Eq. (2.1) involves the kinetic energy of paramagnetic muonium (muon plus an electron) K_{Mu} and its associated spin Hamiltonian $H_{\mathrm{Mu}}^{\mathrm{sp}}$. The gas Hamiltonian H_G' associated with muonium is that for $N-1$ gas atoms plus one positive ion. This positive ion is assumed to be lost in the bath gas, that is, it does not collide with either the bare muon or muonium after its formation. Finally, $V_{i\mathrm{Mu}}$ is the translational potential between muonium and a neutral gas atom. The collisions act as sources and sinks of muons and muonium in this problem. They are assumed to have no direct effect on the spin dynamics.

Associated with the total Hamiltonain is the Liouville superoperator, $\mathscr{L} = \hbar^{-1}[H_T, \ldots]_-$, which generates the dynamics of the full $(N+1)$-particle density operator, that is,

$$d\rho_T(t)/dt = -i\mathscr{L}_T\rho_T(t)$$
$$= -i\left[\mathscr{K}_\mu + \mathscr{L}_\mu^{\rm sp} + \mathscr{L}_G + \sum_{i=1}^{N}\mathscr{V}_{i\mu}\right]\rho_T(t)$$
$$= -i\left[\mathscr{K}_{\rm Mu} + \mathscr{L}_{\rm Mu}^{\rm sp} + \mathscr{L}'_G + \sum_{i=1}^{N}\mathscr{V}_{i\rm Mu}\right]\rho_T(t)\ . \qquad (2.2)$$

The script quantities are the appropriate Liouville superoperators for the various Hamiltonians, that is $\hbar^{-1}$ times the commutator with that Hamiltonian.

Not all the information contained in the full density operator is required for the description of the spin dynamics. Indeed, it is only a small subset that is needed. In particular, the single-particle reduced density operators for muonium,

$$\rho_{\rm Mu}(t) = \mathscr{P}_{\rm Mu}\rho_T(t) = {\rm Tr}_G^{\rm Mu}\rho_T(t)/N!\ , \qquad (2.3)$$

and for the bare muon,

$$\rho_\mu(t) = \mathscr{P}_\mu\rho_T(t) = 1_e^{\rm sp}\otimes{\rm Tr}_G^\mu\rho_T(t)/N!\ , \qquad (2.4)$$

play a crucial role in Boltzmann kinetics. The unit operator for the spin degree of freedom of an electron is explicitly included for the bare muon so that both these density operators have the same number of degrees of freedom, namely, five. That is, three translational degrees of freedom and two spin degrees. Applying the appropriate projection superoperators to Eq. (2.2) leads to the following pair of coupled first-order differential equations:

$$d\rho_\mu(t)/dt + i[\mathscr{K}_\mu + \mathscr{L}_\mu^{\rm sp}]\rho_\mu(t) = -i\,{\rm Tr}_1^\mu\mathscr{V}_{1\mu}\rho_{\mu 1}^{(2)}(t)\ , \qquad (2.5)$$

$$d\rho_{\rm Mu}(t)/dt + i[\mathscr{K}_{\rm Mu} + \mathscr{L}_{\rm Mu}^{\rm sp}]\rho_{\rm Mu}(t) = -i\,{\rm Tr}_1^{\rm Mu}\mathscr{V}_{1\rm Mu}\rho_{\rm Mu1}^{(2)}(t)\ . \qquad (2.6)$$

The trace in Eq. (2.5) is over both the translational degrees of freedom of the gas, denoted by 1, and all but one of the electronic spin degrees of freedom. On the other hand, the trace in Eq. (2.6) is over all translational and all spin degrees of freedom of the gas atom. Thus, both equations retain the same dimensionality. These first-order BBGKY equations[17] involve the two-particle reduced density operators

$$\rho_{\alpha 1}^{(2)}(t) = {\rm Tr}_{N-1}^\alpha\rho_T(t)/(N-1)!\ , \qquad (2.7)$$

where α is either the bare muon or muonium.

Equations (2.5) and (2.6) are, of course, exact. However, they are not closed as they involve a pair of two-particle reduced density operators. To close this system of equations the standard procedure is to apply the generalized Boltzmann ansatz[17] wherein the two-particle reduced density operators are replaced by the product of the appropriate Møller superoperator[18–20] with the appropriate pair of single-particle reduced density operators, namely,

$$\rho_{\alpha 1}^{(2)}(t) = \Omega_{L,\mu 1}\rho_1(t)\rho_\mu(t) + \Omega_{L,\rm Mu1}\rho_1(t)\rho_{\rm Mu}(t)\ . \qquad (2.8)$$

Here, the single-particle reduced gas atom density operator is

$$\rho_1(t) = {\rm Tr}_{N-1}{\rm Tr}_\alpha\rho_T(t)/(N-1)!\ , \qquad (2.9)$$

where again α is either the muon or muonium. Also, Eq. (2.8) involves the Møller superoperator[21]

$$\Omega_{L,\alpha 1} = \lim_{t\to-\infty}\exp(i\mathscr{L}_{\alpha 1}t)\exp[-i(\mathscr{K}_\alpha + \mathscr{K}_1)t]\ , \qquad (2.10)$$

which describes a single binary collision between a gas atom and either a bare muon or muonium. This collision superoperator contains all possible dynamical outcomes of such a single collision which includes elastic, inelastic, and reactive collisions. Here, $\mathscr{K}_1$ is the kinetic Liouville superoperator for the gas atom while $\mathscr{L}_{\alpha 1}$ is the Liouville superoperator associated with the Hamiltonian $H_{\alpha 1} = K_\alpha + K_1 + V_{1\alpha}$. It is to be stressed that these collisions are independent of the spin degrees of freedom. Applying Eq. (2.8) to Eqs. (2.5) and (2.6) leads to the following pair of first-order linear coupled Boltzmann equations for the translational and spin degrees of freedom of the bare muon and muonium:

$$d\rho_\mu(t)/dt + i[\mathscr{K}_\mu + \mathscr{L}_\mu^{\rm sp}]\rho_\mu(t) = \mathscr{C}_{\mu,\mu}(t)\rho_\mu(t) + \mathscr{C}_{\mu,\rm Mu}(t)\rho_{\rm Mu}(t)\ , \qquad (2.11)$$

$$d\rho_{\rm Mu}(t)/dt + i[\mathscr{K}_{\rm Mu} + \mathscr{L}_{\rm Mu}^{\rm sp}]\rho_{\rm Mu}(t) = \mathscr{C}_{\rm Mu,Mu}(t)\rho_{\rm Mu}(t) + \mathscr{C}_{{\rm Mu},\mu}(t)\rho_\mu(t)\ .$$

These equations involve four time-dependent linear Boltzmann collision superoperators. They have the following form:

$$\mathscr{C}_{\alpha,\beta}(t) = -i\,{\rm Tr}_1^\alpha\mathscr{P}_\alpha\mathscr{T}_{\alpha\beta}\rho_1(t)\ . \qquad (2.12)$$

Such a superoperator describes a single binary scattering event wherein a collision between a muon of species β and a gas atom results in the muon being left in the species α. Here

$$\mathscr{T}_{\alpha\beta} = \mathscr{V}_{\alpha 1}\Omega_{L,\beta 1} \qquad (2.13)$$

is the transition superoperator[21] for collisions beginning with β and ending with α. The time dependence of these Boltzmann collision operators is determined by the time dependence of the single-particle reduced density operator for a gas atom. This latter density operator satisfies the standard nonlinear Boltzmann equation for a test gas particle in a bulk gas. However, for practical purposes, the moderating gas can be considered as being homogeneous and at rest. Thus, this single-particle nonequilibrium reduced density operator can be replaced with the usual Maxwell-Boltzmann equilibrium density operator as is done in Sec. III.

Since the observable change of interest is the muon spin vector then the explicit time dependence of the translational degrees of freedom of the single-particle reduced density operators are not required. Thus, these degrees of freedom can be traced over. To do so it is convenient to assume that the single-particle reduced density operators can be factored into a translational density operator and a spin density operator, namely,

$$\rho_\alpha(t)=\rho_\alpha^{\rm tr}(t)\otimes\rho_\alpha^{\rm sp}(t) . \tag{2.14}$$

This seems reasonable since there is no direct coupling between the translational and spin degrees of freedom. Using Eq. (2.14) and tracing over the translational degrees of freedom leads to the following pair of first-order linear differential equations for the spin degrees of freedom of the muon and muonium:

$$d\rho_\mu^{\rm sp}(t)/dt+i\mathscr{L}_\mu^{\rm sp}\rho_\mu^{\rm sp}(t) = -\mathscr{R}_{\mu,\mu}(t)\rho_\mu^{\rm sp}(t)+\mathscr{R}_{\mu,{\rm Mu}}(t)\rho_{\rm Mu}^{\rm sp}(t) , \tag{2.15}$$

$$d\rho_{\rm Mu}^{\rm sp}(t)/dt+i\mathscr{L}_{\rm Mu}^{\rm sp}\rho_{\rm Mu}^{\rm sp}(t) = -\mathscr{R}_{{\rm Mu},{\rm Mu}}(t)\rho_{\rm Mu}^{\rm sp}(t)+\mathscr{R}_{{\rm Mu},\mu}(t)\rho_\mu^{\rm sp}(t) .$$

These equations involve time-dependent rates, namely,

$$\mathscr{R}_{\alpha,\beta}(t)={\rm Tr}_\alpha^{\rm tr}{\rm Tr}_1^\alpha\mathscr{P}_\alpha(\pm i)\mathscr{T}_{\alpha\beta}\rho_1(t)\rho_\beta^{\rm tr}(t)\mathscr{P}_\beta^{\rm sp} , \tag{2.16}$$

where the plus sign is associated with the α,α rates while the minus sign goes with the α,β rates. When α is equal to β the rates describe the probability that the single collision results in the loss of the α state muon whereas when α is not equal to β the rate describes a gain of state α. These rates are related to the number density of the moderating gas and the total cross sections for the appropriate process in Sec. III.

The rate equations (2.15) describe the spin dynamics of the diamagnetic muon and paramagnetic muonium species which are coupled by the charge-exchange cycles that interconvert muon to muonium in the charge-exchange region. These equations form a closed set. However, the rates involve the time-dependent single-particle reduced density operators for a gas atom and the muon. These latter density operators can be expressed in terms of translational Boltzmann equations. Thus, the spin dynamics require the solution of the translational motion problem as well. Solutions of this pair of equations, evaluated at the end of the charge-exchange regime, are the initial conditions for the observable thermal spin polarization.

B. Interaction picture

The rate equations have been derived using the Schrödinger picture. However, such a description does not explicitly depict the sequence of single binary collisions which is inherent in this kinetic theory. An interaction picture, namely,

$$\bar\rho_\alpha^{\rm sp}(t)=\mathscr{G}_\alpha(0,t)\rho_\alpha^{\rm sp}(t) , \tag{2.17}$$

is now introduced to explicitly depict this aspect. Equation (2.17) involves the motion group

$$\mathscr{G}_\alpha(t,0)=T\exp\left[-\int_0^t ds[i\mathscr{L}_\alpha^{\rm sp}+\mathscr{R}_{\alpha,\alpha}(s)]\right] , \tag{2.18}$$

which contains the free-flight spin dynamics generated by $\mathscr{L}_\alpha^{\rm sp}$ along with the probability per single collision that the reaction α to β has occurred. Thus, this group describes the survival probability and spin dynamics of the α state of the muon from time zero to time t.

The diamagnetic muon and paramagnetic muonium interaction-picture single-particle reduced density operators satisfy the following pair of coupled equations:

$$\begin{aligned} d\bar\rho_\mu^{\rm sp}(t)/dt&=\bar{\mathscr{R}}_{\mu,{\rm Mu}}(t)\bar\rho_{\rm Mu}^{\rm sp}(t) ,\\ d\bar\rho_{\rm Mu}^{\rm sp}(t)/dt&=\bar{\mathscr{R}}_{{\rm Mu},\mu}(t)\bar\rho_\mu^{\rm sp}(t) . \end{aligned} \tag{2.19}$$

Here the dynamics is generated by the interaction-picture rates

$$\bar{\mathscr{R}}_{\alpha,\beta}(t)=\mathscr{G}_\alpha(0,t)\mathscr{R}_{\alpha,\beta}(t)\mathscr{G}_\beta(t,0) , \tag{2.20}$$

which describe the coupling of the motions in the α and β states. That is, the free-flight dynamics in state α is followed from time zero to t at which point a transition to state β occurs. This transition is determined by the rate superoperator $\mathscr{R}_{\alpha,\beta}(t)$ which describes the single collision gain of state of β from the scattering of state α and a gas atom. Finally, free-flight motion in state β is followed backward from time t to zero. Using the formal time-ordered solutions of Eqs. (2.19) the Schrödinger-picture density operators become

$$\begin{aligned}\rho_\mu^{\rm sp}(t)=\sum_{\alpha=0}^{\infty}\int_0^t ds_1\int_0^{s_1}ds_2\cdots\int_0^{s_{2\alpha-2}}ds_{2\alpha-1}\int_0^{s_{2\alpha-1}}ds_{2\alpha}\,\mathscr{G}_\mu(t,s_1)\mathscr{R}_{\mu,{\rm Mu}}(s_1)\mathscr{G}_{\rm Mu}(s_1,s_2)\mathscr{R}_{{\rm Mu},\mu}(s_2)\cdots\\ \times\mathscr{G}_{\rm Mu}(s_{2\alpha-1},s_{2\alpha})\mathscr{R}_{{\rm Mu},\mu}(s_{2\alpha})\mathscr{G}_\mu(s_{2\alpha},0)\rho_\mu^{\rm sp}(0) ,\end{aligned}$$

$$\begin{aligned}\rho_{\rm Mu}^{\rm sp}(t)=\sum_{\alpha=0}^{\infty}\int_0^t ds_1\int_0^{s_1}ds_2\cdots\int_0^{s_{2\alpha-1}}ds_{2\alpha}\int_0^{s_{2\alpha}}ds_{2\alpha+1}\,\mathscr{G}_{\rm Mu}(t,s_1)\mathscr{R}_{{\rm Mu},\mu}(s_1)\mathscr{G}_\mu(s_1,s_2)\mathscr{R}_{\mu,{\rm Mu}}(s_2)\cdots\\ \times\mathscr{G}_{\rm Mu}(s_{2\alpha},s_{2\alpha+1})\mathscr{R}_{{\rm Mu},\mu}(s_{2\alpha+1})\mathscr{G}_\mu(s_{2\alpha+1},0)\rho_\mu^{\rm sp}(0) .\end{aligned} \tag{2.21}$$

These solutions clearly depict the sequential collision nature of the Boltzmann equations described by the Schrödinger-picture single-particle reduced density operators for the diamagnetic and paramagnetic species of the muon. Differentiation of Eqs. (2.21) with respect to time reproduce Eqs. (2.15). Thus, any exact solution of the Scrhödinger-picture rate equations contains this sequential collisions aspect implicitly. In Sec IV these formal solutions are related to the observed experimental signal that is seen in the thermal regime while, in Sec. III, the rate superoperators are considered in detail.

III. TIME-DEPENDENT RATES

The rate superoperators, Eq. (2.16), involve the single-particle gas atom reduced density operator which can be obtained by solving the standard nonlinear Boltzmann equation for a test gas atom in the bulk gas at thermal equilibrium. However, since deviations from equilibrium are expected to be small, this single-particle gas atom reduced density operator is taken to be the equilibrium value. That is, it is the product

$$\rho_1(t)=\rho_{1,\mathrm{eq}}^{\mathrm{tr}}\otimes\prod_e\rho_{e,\mathrm{eq}}^{\mathrm{sp}} \tag{3.1}$$

of equilibrium electron-spin density operators with the translational density

$$\rho_{1,\mathrm{eq}}^{\mathrm{tr}}=\int d\vec{r}_1 d\vec{p}_1\,|\,\vec{r}_1,\vec{p}_1\rangle\rangle_{\mathscr{S}}\,n_1 f_1(\vec{p}_1)\ . \tag{3.2}$$

Here $|\,\vec{r},\vec{p}\,\rangle\rangle_{\mathscr{S}}$ and $|\,\vec{r},\vec{p}\,\rangle\rangle_{\mathscr{O}}$ are phase-space representation elements.[22] This latter density operator involves the number density n_1 of the gas and the Maxwell-Boltzmann distribution[14] for the atom's momentum. Thus, the rate superoperator,

$$\mathscr{R}_{\alpha,\mathrm{Mu}}(t)=R_{\alpha,\mathrm{Mu}}(t)\mathscr{P}_{\mathrm{Mu}}^{\mathrm{sp}}\ , \tag{3.3}$$

is the product of a collision rate function

$$R_{\alpha,\mathrm{Mu}}(t)=\mathrm{Tr}_\alpha^{\mathrm{tr}}\mathrm{Tr}_1^{\mathrm{tr}}\mathscr{P}_\alpha(\pm i)\mathscr{T}_{\alpha\mathrm{Mu}}\rho_{1,\mathrm{eq}}^{\mathrm{tr}}\rho_{\mathrm{Mu}}^{\mathrm{tr}}(t)$$
$$=n_1K_{\alpha,\mathrm{Mu}}(t) \tag{3.4}$$

and the projection superoperator on to the paramagnetic spin state while the rate

$$\mathscr{R}_{\alpha,\mu}(t)=R_{\alpha,\mu}(t)\rho_{e,\mathrm{eq}}^{\mathrm{sp}}\mathscr{P}_\mu^{\mathrm{sp}} \tag{3.5}$$

is the product of a collision rate function

$$R_{\alpha,\mu}(t)=\mathrm{Tr}_\alpha^{\mathrm{tr}}\mathrm{Tr}_\mu^{\mathrm{tr}}\mathscr{P}_\alpha(\pm i)\mathscr{T}_{\alpha\mu}\rho_{1,\mathrm{eq}}^{\mathrm{tr}}\rho_\mu^{\mathrm{tr}}(t)$$
$$=n_1K_{\alpha,\mu}(t) \tag{3.6}$$

and an equilibrium electron-spin density operator and the projection superoperator on to the diamagnetic muon spin state. Again the plus sign occurs when α is equal to either μ or Mu while the minus sign appears when they are not equal. That is, the former is a loss term while the latter is a gain. These rate functions involve the number density of the gas and time-dependent rate constants, namely,

$$K_{\alpha\beta}(t)=\int d\vec{r}_\alpha d\vec{p}_\alpha d\vec{r}_1 d\vec{p}_1\int d\vec{r}_\beta d\vec{p}_\beta d\vec{r}\,'_1 d\vec{p}\,'_1\ {}_{\mathscr{O}}\langle\langle\,\vec{r}_\alpha,\vec{p}_\alpha;\vec{r}_1,\vec{p}_1\,|\,\mathscr{P}_\alpha(\pm i)\mathscr{T}_{\alpha\beta}\,|\,\vec{r}_\beta,\vec{p}_\beta;\vec{r}\,'_1,\vec{p}\,'_1\,\rangle\rangle_{\mathscr{S}}$$
$$\times f_1(\vec{p}_1)f_\beta^{\mathrm{tr}}[\vec{r}_\beta,\vec{p}_\beta\,|\,t]\ . \tag{3.7}$$

These latter functions are constant with respect to the concentration of gas atoms, but are dependent upon time through the translational density operator for state β. In center of mass and relative coordinates the transition superoperator becomes

$${}_{\mathscr{O}}\langle\langle\,\vec{r}_\alpha,\vec{p}_\alpha;\vec{r}_1,\vec{p}_1\,|\,\mathscr{P}_\alpha(\pm i)\mathscr{T}_{\alpha\beta}\,|\,\vec{r}_\beta,\vec{p}_\beta;\vec{r}\,'_1,\vec{p}\,'_1\,\rangle\rangle_{\mathscr{S}}$$
$$=\delta(\vec{R}_{\alpha 1}^{\mathrm{c.m.}}-\vec{R}_{\beta 1}^{\mathrm{c.m.}})\delta(\vec{P}_{\alpha 1}^{\mathrm{c.m.}}-\vec{P}_{\beta 1}^{\mathrm{c.m.}})\ {}_{\mathscr{O}}\langle\langle\,\vec{r}_{\alpha 1}^{\,\mathrm{rel}},\vec{p}_{\alpha 1}^{\,\mathrm{rel}}\,|\,\mathscr{P}_\alpha(\pm i)\mathscr{T}_{\alpha\beta}\,|\,\vec{r}_{\beta 1}^{\,\mathrm{rel}},\vec{p}_{\beta 1}^{\,\mathrm{rel}}\rangle\rangle_{\mathscr{S}}\ . \tag{3.8}$$

Thus, the time-dependent rate constant becomes

$$K_{\alpha\beta}(t)=\int d\vec{p}_{\beta 1}^{\,\mathrm{c.m.}}d\vec{p}_{\beta 1}^{\,\mathrm{rel}}\int d\hat{p}_{\alpha 1}^{\mathrm{rel}}\sigma_{\mathrm{gen}}(\beta\vec{p}_{\beta 1}^{\,\mathrm{rel}}\to\alpha\hat{p}_{\alpha 1}^{\,\mathrm{rel}})(p_{\beta 1}^{\mathrm{rel}}/m_{\beta 1})f_1[M_1\vec{p}_{\beta 1}^{\,\mathrm{c.m.}}/M_{\beta 1}-\vec{p}_{\beta 1}^{\,\mathrm{rel}}]$$
$$\times\bar{f}_\beta[M_\beta\vec{p}_{\beta 1}^{\,\mathrm{c.m.}}/M_{\beta 1}+\vec{p}_{\beta 1}^{\,\mathrm{rel}}\,|\,t]\ , \tag{3.9}$$

where

$$\bar{f}_\beta[\vec{p}\,|\,t]=\int d\vec{r}f_\beta[\vec{r},\vec{p}\,|\,t] \tag{3.10}$$

is the momentum Wigner function associated with the β state. Equation (3.9) also involves the generalized cross section[23,24]

$$\sigma_{\mathrm{gen}}(\beta\vec{p}_{\beta 1}^{\,\mathrm{rel}}\to\alpha\hat{p}_{\alpha 1}^{\,\mathrm{rel}})=(-im_{\beta 1}/p_{\beta 1}^{\mathrm{rel}})\int d\vec{r}_{\alpha 1}^{\,\mathrm{rel}}d\vec{r}_{\beta 1}^{\,\mathrm{rel}}\int_0^\infty dp_{\alpha 1}^{\mathrm{rel}}(p_{\alpha 1}^{\mathrm{rel}})^2\ {}_{\mathscr{O}}\langle\langle\,\vec{r}_{\alpha 1}^{\,\mathrm{rel}},\vec{p}_{\alpha 1}^{\,\mathrm{rel}}\,|\,\mathscr{P}_\alpha\mathscr{T}_{\alpha\beta}\,|\,\vec{r}_{\beta 1}^{\,\mathrm{rel}},\vec{p}_{\beta 1}^{\,\mathrm{rel}}\rangle\rangle_{\mathscr{S}}$$
$$=\sigma(\beta\vec{p}_{\beta 1}^{\,\mathrm{rel}}\to\alpha\hat{p}_{\alpha 1}^{\,\mathrm{rel}})-\delta_{\alpha\beta}\delta(\hat{p}_{\alpha 1}^{\,\mathrm{rel}}-\hat{p}_{\beta 1}^{\,\mathrm{rel}})\sigma_{\mathrm{tot}}(\beta)\ , \tag{3.11}$$

where

$$\sigma(\beta\vec{p}_{\beta 1}^{\,\mathrm{rel}}\to\alpha\hat{p}_{\alpha 1}^{\,\mathrm{rel}})$$
$$=(2\pi\hbar)^4(p_{\alpha 1}^{\mathrm{rel}})\,|\,\langle\alpha\vec{p}_{\alpha 1}^{\,\mathrm{rel}}\,|\,P_\alpha t_{\alpha\beta}\,|\,\beta\vec{p}_{\beta 1}^{\,\mathrm{rel}}\rangle\,|^2/p_{\beta 1}^{\mathrm{rel}} \tag{3.12}$$

is the differential cross section[25] for the transition from α to β and where

$$\sigma_{\mathrm{tot}}(\beta)=\mathrm{Im}[-(4\pi h^2 m_{\beta 1})\langle\beta\vec{p}_{\beta 1}^{\,\mathrm{rel}}\,|\,t_{\beta\beta}\,|\,\beta\vec{p}_{\beta 1}^{\,\mathrm{rel}}\rangle/p_{\beta 1}^{\mathrm{rel}}]$$
$$=\sigma_{\mathrm{tot}}(\beta\to\beta)+\sigma_{\mathrm{tot}}(\beta\to\alpha) \tag{3.13}$$

is the associated total cross section.[25] The last line of Eq. (3.13) involves the cross sections for β to β events and β to α events. Since generalized cross sections conserve particles[23,24] then the time-dependent rate constants become

$$K_{\alpha\beta}(t)=\int d\vec{p}\,^{\text{c.m.}}_{\beta 1}d\vec{p}\,^{\text{rel}}_{\beta 1}\sigma_{\text{tot}}(\beta\to\alpha)(p^{\text{rel}}_{\beta 1}/m_{\beta 1})$$
$$\times f_1[M_1\vec{p}\,^{\text{c.m.}}_{\beta 1}/M_{\beta 1}-\vec{p}\,^{\text{rel}}_{\beta 1}]$$
$$\times f_\beta[M_\beta\vec{p}\,^{\text{c.m.}}_{\beta 1}/M_{\beta 1}+\vec{p}\,^{\text{rel}}_{\beta 1}\,|\,t]$$
$$=K_{\beta\beta}(t)\ . \tag{3.14}$$

For β equal to Mu the rate constant $K_{\alpha\beta}(t)$ is now denoted as $K_L(t)$ while for β equal to μ it is $K_C(t)$, that is, they are the time-dependent rate constants for electron loss and capture, respectively. Making use of Eq. (3.14) the rate equations (2.15) become

$$\begin{aligned}
&d\rho^{\text{sp}}_\mu(t)/dt+i\mathscr{L}^{\text{sp}}_\mu\rho^{\text{sp}}_\mu(t)\\
&\quad=[-K_C(t)\rho^{\text{sp}}_{e,\text{eq}}\rho^{\text{sp}}_\mu(t)+K_L(t)\rho^{\text{sp}}_{\text{Mu}}(t)]n_1\ ,\\
&d\rho^{\text{sp}}_{\text{Mu}}(t)/dt+i\mathscr{L}^{\text{sp}}_{\text{Mu}}\rho^{\text{sp}}_{\text{Mu}}(t)\\
&\quad=[-K_L(t)\rho^{\text{sp}}_{\text{Mu}}(t)+K_C(t)\rho^{\text{sp}}_{e,\text{eq}}\rho^{\text{sp}}_\mu(t)]n_1\ .
\end{aligned}\tag{3.15}$$

Solutions of Eqs. (3.15) describe the effects that the charge-exchange regime has on the spin dynamics. Such solutions indirectly involve the translational motion through the time-dependent rate constants. Thus, for purposes of solving Eqs. (3.15), these rates can be considered as parameters which may be extracted from experiments or calculated from scattering theory. The relation between these solutions and the observed μSR signals are developed in Sec. IV.

IV. RELATION TO OBSERVED SIGNAL

In the thermalization and thermal regimes the density operator which describes the spin dynamics has the form

$$\begin{aligned}\rho_S(t)=&\exp[-i\mathscr{L}^{\text{sp}}_\mu(t-t_2)]\rho^{\text{sp}}_\mu(t_2)\\
&+\exp[-i\mathscr{L}^{\text{sp}}_{\text{Mu}}(t-t_2)]\rho^{\text{sp}}_{\text{Mu}}(t_2)\ ,\end{aligned}\tag{4.1}$$

where $\rho^{\text{sp}}_\mu(t_2)$ and $\rho^{\text{sp}}_{\text{Mu}}(t_2)$ are solutions of Eqs. (3.15) evaluated at the end (t_2) of the cyclic charge-exchange region. These final density operators for the charge-exchange region act as the initial conditions for the subsequent thermalization and thermal regimes. They are now related to the amplitudes and phases of the observed experimental signals. Since only the muon spin polarization is observed, it is then convenient to trace over the electronic spin, namely,

$$\rho_I(t)=\text{Tr}_e\rho_S(t)=\rho^\mu_I(t)+\rho^{\text{Mu}}_I(t)\ . \tag{4.2}$$

The resulting effective muon spin density operator consists of two contributions: one from the diagmagnetic term

$$\rho^\mu_I(t)=\exp[-i\mathscr{L}^{\text{sp}}_\mu(t-t_2)]\text{Tr}_e\rho^{\text{sp}}_\mu(t_2) \tag{4.3}$$

and one from the paramagnetic term

$$\rho^{\text{Mu}}_I(t)=\text{Tr}_e\exp[-i\mathscr{L}^{\text{sp}}_{\text{Mu}}(t-t_2)]\rho^{\text{sp}}_{\text{Mu}}(t_2)\ . \tag{4.4}$$

The motion of the former is generated by the diamagnetic muon Liouville superoperator which is $\hbar^{-1}$ times the commutator with the muon spin Hamiltonian

$$H^{\text{sp}}_\mu=-\omega_\mu\vec{\text{I}}\cdot\hat{Z},\ \ \omega_\mu=g_\mu\beta_\mu B/\hbar=8.6\times 10^4 B\ , \tag{4.5}$$

in units of sec^{-1}, where there is an external magnetic field in the $\hat{Z}$ direction. Here ω_μ is the diamagnetic muon Larmor frequency while g_μ is the muon g factor and $\beta_\mu=|e|\hbar/2m_\mu c$ is the muon Bohr magneton. The motion of the latter density operator is generated by the paramagnetic muonium Liouville superoperator which is $\hbar^{-1}$ times the commutator with the muonium spin Hamiltonian[1]

$$H^{\text{sp}}_{\text{Mu}}=\omega_e\vec{\text{S}}\cdot\hat{Z}-\omega_\mu\vec{\text{I}}\cdot\hat{Z}+\hbar^{-1}\omega_0\vec{\text{I}}\cdot\vec{\text{S}}\ . \tag{4.6}$$

Here ω_e is the electronic Larmor frequency while ω_0 is the muonium hyperfine frequency, 2.8×10^{10} rad/sec.

Making use of Eq. (4.2) the observed polarization $P_X=(2/\hbar)\vec{\text{I}}\cdot\hat{X}$ at time t $(>t_2)$

$$P_X(t)=\text{Tr}_\mu P_X\rho_I(t)=P^\mu_X(t)+P^{\text{Mu}}_X(t)\ , \tag{4.7}$$

becomes the sum of two terms. To evaluate these contributions to the polarization it is convenient to use a particular representation. The representation used in the following is that of the eigenfunctions[1] of the muonium spin Hamiltonian, namely,

$$\begin{aligned}
&|1\rangle=|\alpha\alpha\rangle\ ,\ \ E_1=\hbar(\omega_{\text{Mu}}+\omega_0/4)\ ,\\
&|2\rangle=s\,|\alpha\beta\rangle+c\,|\beta\alpha\rangle\ ,\\
&E_2=-(\hbar\omega_0/2)[\tfrac{1}{2}-(1+x^2)^{1/2}]\ ,\\
&|3\rangle=|\beta\beta\rangle, E_3=\hbar(-\omega_{\text{Mu}}+\omega_0/4),\\
&|4\rangle=c\,|\alpha\beta\rangle-s\,|\beta\alpha\rangle\ ,\\
&E_4=-(\hbar\omega_0/2)[\tfrac{1}{2}+(1+x^2)^{1/2}]\ ,
\end{aligned}\tag{4.8}$$

where

$$\begin{aligned}\omega_{\text{Mu}}=(\omega_e-\omega_\mu)/2&=\omega_e(1-m_e/m_\mu)/2\\
&=8.8\times 10^6 B\\
&=(m_\mu/m_e-1)\omega_\mu/2=103\omega_\mu\ ,\end{aligned}\tag{4.9}$$

in units of sec^{-1}, is the muonium Larmor frequency. Equations (4.8) also involve a field strength parameter

$$\begin{aligned}x=(\omega_e+\omega_\mu)/\omega_0&=(\omega_e/\omega_0)(1-m_e/m_\mu)\\
&=6.3\times 10^{-4}B\end{aligned}\tag{4.10}$$

and a pair of normalization constants

$$c=[1+x/(1+x^2)^{1/2}]^{1/2}/2^{1/2}\ ,\ \ s=(1-c^2)^{1/2}\ . \tag{4.11}$$

The original two spin basis can be reexpressed in terms of this muonium basis, that is,

$$\begin{aligned}
&|\alpha\alpha\rangle=|1\rangle\ ,\\
&|\alpha\beta\rangle=s\,|2\rangle+c\,|4\rangle\ ,\\
&|\beta\beta\rangle=|3\rangle\ ,\\
&|\beta\alpha\rangle=c\,|2\rangle-s\,|4\rangle\ .
\end{aligned}\tag{4.12}$$

Making use of this basis the diamagnetic contribution to the observed spin polarization

$$P_X^{\mu}(t)=\mathrm{Tr}_{\mu}P_X\rho_I^{\mu}(t)=2\,\mathrm{Re}\rho_{\alpha\beta}^{\mu}(t)$$
$$=P_{\mu}\cos(\omega_{\mu}t+\theta_{\mu})\ , \tag{4.13}$$

involves an amplitude

$$P_{\mu}=|\rho_{\alpha\beta}^{\mu}(t_2)|=|c[\rho_{12}^{\mu}(t_2)+\rho_{43}^{\mu}(t_2)]+s[\rho_{23}^{\mu}(t_2)-\rho_{14}^{\mu}(t_2)]|\ ,$$

and a phase

$$\theta_{\mu}=-\omega_{\mu}t_2+\theta_{\alpha\beta}^{\mu}(t_2)\ . \tag{4.14}$$

Equations (4.14) relate the observed experimental amplitude and phase associated with the diamagnetic signal to the diamagnetic density operator that emerges from the charge-exchange region. On the other hand, the paramagnetic contribution consists of four parts, namely,

$$\begin{aligned}P_X^{\mathrm{Mu}}(t)&=\mathrm{Tr}_{\mu}P_X\rho_I^{\mathrm{Mu}}(t)\\&=\mathrm{Tr}_{\mu}P_X\left[\mathrm{Tr}_e\sum_{ij}|i,j\rangle\rangle\rho_{ij}^{\mathrm{Mu}}(t_2)\exp[-i(\omega_i-\omega_j)(t-t_2)]\right]\\&=P_{\mathrm{Mu}}^1\cos[(\omega_{\mathrm{Mu}}+\Omega)t-\theta_{\mathrm{Mu}}^1]+P_{\mathrm{Mu}}^2\cos[(\omega_{\mathrm{Mu}}-\Omega)t-\theta_{\mathrm{Mu}}^2]+P_{\mathrm{Mu}}^3\cos[(\omega_0+\omega_{\mathrm{Mu}}+\Omega)t-\theta_{\mathrm{Mu}}^3]\\&\quad+P_{\mathrm{Mu}}^4\cos[(\omega_0-\omega_{\mathrm{Mu}}+\Omega)t-\theta_{\mathrm{Mu}}^4]\ .\end{aligned} \tag{4.15}$$

These terms in the polarization are related to the following components of the muonium part of the density operator:

$$\begin{aligned}&P_{\mathrm{Mu}}^1=|2s\rho_{23}^{\mathrm{Mu}}(t_2)|\ ,\quad \theta_{\mathrm{Mu}}^1=(\omega_{\mathrm{Mu}}+\Omega)t_2+\theta_{23}^{\mathrm{Mu}}(t_2)\ ,\\&P_{\mathrm{Mu}}^2=|2c\rho_{12}^{\mathrm{Mu}}(t_2)|\ ,\quad \theta_{\mathrm{Mu}}^2=(\omega_{\mathrm{Mu}}-\Omega)t_2+\theta_{12}^{\mathrm{Mu}}(t_2)\ .\\&P_{\mathrm{Mu}}^3=|-2s\rho_{14}^{\mathrm{Mu}}(t_2)|\ ,\\&\theta_{\mathrm{Mu}}^3=(\omega_0+\omega_{\mathrm{Mu}}+\Omega)t_2+\theta_{14}^{\mathrm{Mu}}(t_2)\ ,\\&P_{\mathrm{Mu}}^4=|2c\rho_{43}^{\mathrm{Mu}}(t_2)|\ ,\\&\theta_{\mathrm{Mu}}^4=(\omega_0-\omega_{\mathrm{Mu}}+\Omega)t_2+\theta_{43}^{\mathrm{Mu}}(t_2)\ .\end{aligned} \tag{4.16}$$

In general, there are five different frequency signals that could be observed experimentally if x is of sufficient size so that

$$\Omega=[(1+x^2)^{1/2}-1]/2 \tag{4.17}$$

is resolvable. However, it is standard experimental practice[1] to measure the muonium signal at about 8 G and the diamagnetic signal at about 70–120 G. As well, the current time resolution for these gas phase experiments is insufficient to observe the muonium hyperfine frequency.[1] Thus, only two signals are currently seen,[1] namely, that associated with the diamagnetic Larmor frequency [see Eq. (4.13)] and that associated with the paramagnetic muonium Larmor frequency, the so-called[1] "triplet" signal. This triplet signal is the Larmor part of the following low-field muonium polarization:

$$P_X^{\mathrm{Mu}}(t)=P_{\mathrm{Mu}}\cos(\omega_{\mathrm{Mu}}t-\theta_{\mathrm{Mu}})+P_{\mathrm{Mu}}^0\cos(\omega_0t-\theta_{\mathrm{Mu}}^0)\ . \tag{4.18}$$

This observable signal also involves the so-called[1] "singlet" signal which is associated with the hyperfine frequency and is, at present, not experimentally resolved. The various amplitudes and phases are given by the following expressions:

$$\begin{aligned}P_{\mathrm{Mu}}&=\left[\left[\sum_{k=1}^{2}P_{\mathrm{Mu}}^k\cos\theta_{\mathrm{Mu}}^k\right]^2+\left[\sum_{k=1}^{2}P_{\mathrm{Mu}}^k\sin\theta_{\mathrm{Mu}}^k\right]^2\right]^{1/2}\\&=\left[\sum_{k=1}^{2}(P_{\mathrm{Mu}}^k)^2+2P_{\mathrm{Mu}}^1P_{\mathrm{Mu}}^2\cos(\theta_{\mathrm{Mu}}^1-\theta_{\mathrm{Mu}}^2)\right]^{1/2}\\&=|2s\rho_{23}^{\mathrm{Mu}}(t_2)+2c\rho_{12}^{\mathrm{Mu}}(t_2)|\\&=[(P_{\mathrm{Mu},R})^2+(P_{\mathrm{Mu},I})^2]^{1/2}\ ,\end{aligned} \tag{4.19}$$

$$\theta_{\mathrm{Mu}}=\arccos(P_{\mathrm{Mu},R}/P_{\mathrm{Mu}})\times\begin{cases}1, & P_{\mathrm{Mu},I}>0\\-1, & P_{\mathrm{Mu},I}<0\end{cases} \tag{4.20}$$

$$\begin{aligned}P_{\mathrm{Mu}}^0&=|-2s\rho_{14}^{\mathrm{Mu}}(t_2)+2c\rho_{43}^{\mathrm{Mu}}(t_2)|\\&=\left[\left[\sum_{k=3}^{4}P_{\mathrm{Mu}}^k\cos\theta_{\mathrm{Mu}}^k\right]^2+\left[\sum_{k=3}^{4}P_{\mathrm{Mu}}^k\sin\theta_{\mathrm{Mu}}^k\right]^2\right]^{1/2}\\&=[(P_{\mathrm{Mu},R}^0)^2+(P_{\mathrm{Mu},I}^0)^2]^{1/2}\ ,\end{aligned} \tag{4.21}$$

and

$$\theta_{\mathrm{Mu}}^0=\arccos(P_{\mathrm{Mu},R}^0/P_{\mathrm{Mu}}^0)\times\begin{cases}1, & P_{\mathrm{Mu},I}^0>0\\-1, & P_{\mathrm{Mu},I}^0<0\ .\end{cases} \tag{4.22}$$

In summary, the experimental signal is given in terms of Eqs. (4.13) and (4.18), namely,

$$P_X^{\mathrm{expt}}(t)=P_{\mu}\cos(\omega_{\mu}t+\theta_{\mu})+P_{\mathrm{Mu}}\cos(\omega_{\mathrm{Mu}}t-\theta_{\mathrm{Mu}})\ . \tag{4.23}$$

V. DISCUSSION

A theoretical framework to describe the spin dynamics associated with the charge-exchange region which occurs during the thermalization of highly energetic muons in gases has been presented. That is, a pair of rate equations (3.15) for the diamagnetic and paramagnetic muon species have been derived from first principles. These equations involve time-dependent rate constants for which explicit formulas have been derived. Relations to the experimentally observed signals have also been presented [Eq. (4.23)]. Direct comparisons with experiments can be made in two ways. One method is to calculate the time-dependent rate constants using scattering theory and to obtain the solutions of the appropriate translational Boltzmann equations. This then gives the rate constants for which solutions of the rate equations can be calculated. The other method, which is applied in the next paper in this series,[13] is to approximate the time-dependent rate constants with time-independent functions. Analytic solutions for the amplitudes and phases of the signals are then obtained. Furthermore, the rate constants and the time span of the region are then considered as parameters in a fitting procedure. The resulting values can then be interpreted in terms of the formal expressions for the rates. This latter point is a subject for future consideration.

This paper has presented a description of the charge-exchange mechanism for the depolarization of the spin of the positive muon as it thermalizes in a dilute gas. In this mechanism the depolarization results from the exchange of polarization between the muon and the electron in muonium mediated by the ground electronic state hyperfine interaction. No spin lattice depolarization effects are involved since they occur after the charge-exchange region in gases. This stands in contrast to condensed phases where it is mainly spin lattice effects rather than the charge-exchange mechanism that leads to depolarization; see, for example, Ref. 26. That is, the time scale of the charge-exchange region is of the order of picoseconds in condensed phases rather than nanoseconds. Such a time scale is too short to allow direct depolarization due to the hyperfine interaction. The rate equations developed here for gases involve time-dependent rates as opposed to the standard phenomenological theory[26] for condensed phase depolarization.

ACKNOWLEDGMENTS

I would like to thank Dr. Masa Senba, Professor Robert Snider, Professor Don Fleming, and Professor Peter Meier for their help in writing this manuscript.

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