2s state, τ_p is the radiative lifetime of the cascading p state, n_p is the p-state density where $n_p = F\rho Q_p \tau_p \times (1 - e^{-y/v\tau_p})$, Q_p is the cross section for producing the p state, and B is the branching ratio which is given by $B = A\tau_p$, where A is the transition probability for $p \rightarrow 2s$. The rate equation for the production of the 2s state outside the cell on the exit side of the cell is Eq. (1) with $\rho = 0$. The solution to these equations for a cell length L gives the density for the 2s atoms at a distance of x from the exit aperture,

$$n_s = F\rho Q\tau_s \left[\left(1 + \frac{BQ_p}{Q(1 - \tau_p/\tau_s)} \right) (1 - e^{-L/v\tau_s}) e^{-x/v\tau_s} - \frac{BQ_p\tau_p/\tau_s (1 - e^{-L/v\tau_p}) e^{-x/v\tau_p}}{Q(1 - \tau_p/\tau_s)} \right] . \quad (2)$$

For $L \ll v\tau_s$ and $v\tau_p \ll x \ll v\tau_s$, Eq. (2) becomes $n_s = F\rho QL\,(1 + BQ_p/Q)/v$. Thus the cascade fraction is just BQ_p/Q.

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Influence of Nuclear Spin on Collisional Relaxation within the Alkali-Metal $^2P_{3/2}$ Excited State*†

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The effect of the hyperfine interaction on collisional relaxation among Zeeman sublevels is evaluated for a $J = \frac{3}{2}$ state coupled to nuclear spins of $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$. Standard depolarization experiments, performed on atoms with hyperfine structure, are discussed in terms of relaxation-probability matrices parametrized in terms of the relative collisional-transition probabilities for a nuclear-spin-zero atom. Experimental results involving D_2 optical pumping of ^{85}Rb and ^{87}Rb are shown to support the approximation of nuclear decoupling in collisional relaxation. The cross sections for electronic collisional relaxation, assuming a van der Waals model for the collisional interaction, are, in units of 10^{-14} cm^2, 1.19 (^{85}Rb-He), 1.09 (^{85}Rb-Ne), 2.44 (^{85}Rb-Ar), 1.13 (^{87}Rb-He), 1.03 (^{87}Rb-Ne), and 2.26 (^{87}Rb-Ar).

INTRODUCTION

The experimental study of collisional relaxation among the Zeeman sublevels of atomic states has roots as far back as Wood's classic experiments on depolarization of resonance radiation.[1] Modern optical-pumping and level-crossing techniques have provided both motivation and means for more recent work. The principal areas of current interest involve measurement of relative probabilities for collisionally induced transitions (relaxation) among the Zeeman sublevels of atomic states, and the extraction of information on the nature of the collisional interaction from such measurements. Experiments have a simple elegance when performed on atomic states possessing electronic

angular momenta of $J=\frac{1}{2}$ or $J=1$, where relaxation rates among the two or three Zeeman sublevels involved can be relatively easily measured and interpreted.[2,3] Complications arise in extensions of work to states of higher angular momenta, and to atoms possessing nuclear spin and hyperfine structure. Relaxation within the $^2P_{3/2}$ first excited states of the alkali-metal atoms, which is particularly interesting from the optical-pumping point of view, presents intriguing complexity through the coupling of these $J=\frac{3}{2}$ states to nuclear spins of $I=\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$.

As far as we are aware, the earliest modern work on collisional relaxation among the Zeeman sublevels of the excited states of alkali-metal atoms, including the effects of the hyperfine interaction, was performed by Bender.[4] Bender introduced a particularly useful approximation, nuclear decoupling, that in many cases allows a separation of the effects of the collisional interaction itself from effects of the hyperfine interaction. In this approximation it is assumed that if the duration of the collision is considerably shorter than the hyperfine period, the direct collisional interaction reorients only the (decoupled) electronic moment. Subsequent reorientation of the nuclear and electronic moments occurs through the hyperfine interaction, provided that the hyperfine period is shorter than the mean time between collisions. Such an approximation has been used with great success to describe collisional relaxation within the $^2S_{1/2}$ alkali-metal ground state[5-9]; it also formed the basis for calculations describing collisional relaxation with the $^2P_{1/2}$ and $^2P_{3/2}$ alkali-metal excited states, subject to the assumption of particular models for the relaxation of $\vec{J}$.[10] In the present work, we extend and generalize the calculations of Ref. 10 to include *any* possible model for the relaxation of $\vec{J}$, coupled to nuclear spins of $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$. We parametrize all calculations in terms of σ, the cross section for electronic ($\vec{J}$) collisional relaxation, that is, the cross section which would be measured in the absence of nuclear spin. We examine in detail significant changes in interpretation of standard depolarization experiments that occur when they are performed on atoms possessing nuclear spin, and we report results from an experiment involving D_2 optical pumping of ^{85}Rb and ^{87}Rb. Similar independent work has been reported recently by Okunevich and Perel,[11] and by Zhitnikov, Kuleshov, Okunevich, and Sevast'yanov.[12]

COLLISIONAL RELAXATION IN $J=\frac{3}{2}$ STATE IN ABSENCE OF NUCLEAR SPIN

It is helpful to begin by reviewing some aspects of collisional relaxation in the absence of hyperfine structure: We assume the nuclear spin ($\vec{I}$) to be zero. We are interested in the particular case of relaxation among the m_J Zeeman sublevels of a $J=\frac{3}{2}$ atomic state induced by collisions with foreign (buffer) gas atoms. We assume that such collisions cause only transitions between m_J sublevels: Transitions out of the J state are assumed not to occur. Examples of systems satisfying this criterion are the $^2P_{3/2}$ first excited states of Rb and Cs, and the $^2P_{3/2}$ ground states of In and Tl suffering collisions with noble-gas atoms.[10] We restrict our discussion to experiments in which only diagonal elements of the density matrix are excited: We therefore shall discuss some common experiments that measure changes of populations of m_J sublevels, but shall not touch upon Hanle-effect or level-crossing work, which involve coherence effects.

In Table I we define the set of relative probabilities for collisionally induced transitions among the four m_J sublevels of a $J=\frac{3}{2}$ atomic state. k_A and k_B represent the relative probabilities of transitions for which the initial and final states are the same, $(m_J \rightarrow m_J',\ m_J = m_J')$. The rest of the k_α represent relative probabilities for transitions between sublevels of different $m_J (m_J \rightarrow m_J',\ m_J \neq m_J')$. The k_α are normalized to unity by

$$k_A + k_1 + k_2 + k_3 = 1 \ , \tag{1a}$$

$$k_B + k_0 + k_1 + k_2 = 1 \ . \tag{1b}$$

The relative transition probabilities k_α are related to the relaxation rates (= transition rates = collision numbers, in other terminology) Z_α by the relation

$$Z_\alpha = k_\alpha \sigma v_{\mathrm{rel}} n_0 p / p_0 \ , \tag{2}$$

where σ is the cross section for collisionally induced electronic relaxation, v_{rel} is the mean relative velocity of an alkali-atom–buffer-gas-atom pair. n_0 is Loschmidt's number, p is the buffer-gas pressure in Torr, and p_0 is 760 Torr.

Using the definitions above, we can write down rate equations describing the relaxation of any given distribution of population throughout the m_J sublevels of the $^2P_{3/2}$ state. If $n(m_J)$ represents the relative population of the m_J sublevel, then, for example,

TABLE I. Definition of relative probabilities for collisionally induced transitions among the m_J sublevels of $J=\frac{3}{2}$ atomic state.

m_J \ m_J	$+\frac{3}{2}$	$+\frac{1}{2}$	$-\frac{1}{2}$	$-\frac{3}{2}$
$+\frac{3}{2}$	k_A	k_1	k_2	k_3
$+\frac{1}{2}$	k_1	k_B	k_0	k_2
$-\frac{1}{2}$	k_2	k_0	k_B	k_1
$-\frac{3}{2}$	k_3	k_2	k_1	k_A

$$\frac{dn_{(3/2)}}{dt} = n\sigma v_{\mathrm{rel}}[(-k_1-k_2-k_3)n_{(3/2)} + k_1 n_{(1/2)} + k_2 n_{(-1/2)} + k_3 n_{(-3/2)}] , \quad (3)$$

where $n=(n_0 p/p_0)$. The set of four equations similar to Eq. (3) yields time-dependent solutions for the $n(m_J)$ that are sums of exponentials:

$$n(m_J) = \sum_i A_i e^{-\gamma_i t} , \quad (4)$$

where the coefficients A_i are determined by the initial distribution of population, and the decay constants γ_i are linear functions of the k_i, and are proportional to σ and v_{rel}.

The set of equations similar to Eq. (3), represents the decay of the diagonal elements of the excited-state density matrix in terms of the decay of populations of individual m_J states. The basis vectors of the system are [1,0,0,0], [0,1,0,0], [0,0,1,0], and [0,0,0,1], where the ordering of m_J states is $(+\frac{3}{2}, +\frac{1}{2}, -\frac{1}{2}, -\frac{3}{2})$. We are not restricted to such a representation. We could just as easily expand a diagonal density matrix in terms of diagonal matrices proportional to [1,1,1,1], [3,1,−1,−3], [1,−1,−1,1], and [1,−3,3,−1]. Such a basis is that of the diagonal components T_0^x, $x=0,1,2,3$, of the irreducible tensor representation. D'Yakanov and Perel[2] and Omont[3] have shown how the irreducible tensor representation can be utilized to effect an enormous simplification of the study of atomic collisional relaxation. In contrast to the multiple-exponential relaxation of individual $n(m_J)$'s, each linear combination of the $n(m_J)$ constituting a T_0^x component decays with a single rate constant Γ_x given by

$$\Gamma_x = \gamma_x \sigma n v_{\mathrm{rel}} . \quad (5)$$

γ_x is the analog, for the T_0^x representation, of k_α. σ, n, and v_{rel} remain as defined in Eq. (2).

Representations of the decay of state populations in terms of the $n(m_J)$ basis or of the T_0^x basis must be equivalent. In the absence of an anisotropic interaction, transformation equations relating the transition probabilities γ_x to the transition probabilities k_α are derived by transforming Eqs. (3) for $dn(m_J)/dt$ to dT_0^x/dt, yielding

$$k_0 = \tfrac{1}{20}(\gamma_1 - 5\gamma_2 + 9\gamma_3) , \quad (6a)$$

$$k_1 = \tfrac{1}{20}(-3\gamma_1 + 5\gamma_2 + 3\gamma_3) , \quad (6b)$$

$$k_2 = \tfrac{1}{20}(3\gamma_1 + 5\gamma_2 + -3\gamma_3) , \quad (6c)$$

$$k_3 = \tfrac{1}{20}(9\gamma_1 - 5\gamma_2 + \gamma_3) . \quad (6d)$$

The γ_x appearing in Eqs. (6a)–(6d) are identical to the γ_i of Eq. (4).[13]

Since we started with six k_α, subject to two normalization equations [Eqs. (1a) and (1b)], one might expect that four independent k_α should remain: Let us take these to be the four k_α for which $m_J \neq m'_J$. In terms of the T_0^x basis γ_0, which represents the collisional decay of the total population of the J state, must be zero to satisfy our initial assumption of no transitions out of the J state. Only three nonzero γ_x remain, i.e., γ_1, γ_2, and γ_3. Thus we are left with four k_α that can be expressed in terms of three γ_x; the four k_α cannot be linearly independent, a fact first pointed out by Gallagher.[14] The equation displaying the dependence of the k_α comes out of the calculation referred to above, and is

$$3k_0 - 4k_1 + 4k_2 - 3k_3 = 0 . \quad (7)$$

We now relate the decay rates Γ_x to experimental observables. The T_0^x components can be represented in terms of angular momentum operators, as indicated in Table II. If the actual number of atoms in the jth sublevel is n_j, the occupation probability of that sublevel is n_j/N, where $N=\sum_j n_j$. We define

$$\langle J_z \rangle = \sum_j n_j \langle J_z \rangle_j / N , \quad (8a)$$

$$\langle J_z^2 \rangle = \sum_j n_j \langle J_z^2 \rangle_j / N , \quad (8b)$$

where $\langle J_z \rangle$ is the dipole moment, or "orientation," and $\langle J_z^2 \rangle$ is the quadrupole moment, or "alignment," of the excited state. It is clear from reference to Table II and the preceding equations that Γ_1, the decay rate of T_0^1, is the decay rate of

TABLE II. Representation of the diagonal irreducible tensor operators for $J=\frac{3}{2}$.

Tensor component	Physical significance	Representation in terms of angular momentum operators	Decay rate
T_0^0	Total population of all m_J states	J^2	$\Gamma_0=0$
T_0^1	"orientation" (dipole moment)	J_z	Γ_1
T_0^2	"alignment" (quadrupole moment)	$3J_z^2 - J^2$	Γ_2
T_0^3	octupole moment	$5J_z^3 + J_z - 3J^2$	Γ_3

TABLE III. Expectation values of J_z and J_z^2 for the m_J sublevels the $^2P_{3/2}$ alkali excited state, with corresponding intensities of Zeeman components of the $D_2(^2P_{3/2} \rightarrow {}^2S_{1/2})$ resonance line ($I=0$).

m_J	$\langle J_z\rangle_i$	$\langle J_z^2\rangle_i$	$(I_{\sigma^+})_i(\parallel H_0)$	$(I_{\sigma^-})_i(\parallel H_0)$	$(I_\sigma)_i(\perp H_0)$	$(I_\pi)_i(\perp H_0)$
$+\frac{3}{2}$	$\frac{3}{2}$	$\frac{9}{1}$	$\frac{1}{2}$	0	$\frac{1}{2}$	0
$+\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{4}$	$\frac{1}{6}$	0	$\frac{1}{6}$	$\frac{2}{3}$
$-\frac{1}{2}$	$-\frac{1}{2}$	$\frac{1}{4}$	0	$\frac{1}{6}$	$\frac{1}{6}$	$\frac{2}{3}$
$-\frac{3}{2}$	$-\frac{3}{2}$	$\frac{9}{1}$	0	$\frac{1}{2}$	$\frac{1}{2}$	0

$\langle J_z\rangle$, and Γ_2, the decay rate of T_0^2, is the decay rate of $\langle J_z^2\rangle$.

Nonzero $\langle J_z\rangle$ can be produced in the $^2P_{3/2}$ state by the absorption of circularly polarized (σ^+ or σ^-) resonance radiation propagating along the direction of the magnetic field (H_0). In equilibrium, and assuming equal populations of ground-state sublevels,

$$\langle J_z\rangle = \tfrac{5}{4}(1+\Gamma_1\tau)^{-1} , \tag{9}$$

where τ is the lifetime of the excited state. $\langle J_z\rangle$ can be monitored by measurement of the degree of circular polarization of the resonance fluorescence, as reference to Table III shows:

$$(I_{\sigma^+}-I_{\sigma^-}) \approx \textstyle\sum_j n_j (I_{\sigma^+}-I_{\sigma^-})_j /N$$
$$= \textstyle\sum_j n_j \langle J_z\rangle_j /3N = \langle J_z\rangle /3 . \tag{10}$$

Measurement of $(I_{\sigma^+}-I_{\sigma^-})$ vs buffer-gas pressure thus yields $(\gamma_1\sigma)$.

Nonzero $\langle J_z^2\rangle$ can be created through the absorption of σ^+ or σ^- light propagating along H_0, or by the absorption of σ or π light propagating in a direction perpendicular to H_0. $\langle J_z^2\rangle$ can be monitored by measuring the degree of linear polarization of the resonance fluorescence emitted perpendicular to H_0:

$$(I_\sigma - I_\pi) \approx \textstyle\sum_j n_j (I_\sigma - I_\pi)_j /N$$
$$= \textstyle\sum_j n_j (\frac{1}{2}\langle J_z^2\rangle - \frac{5}{8})_j /N$$
$$= (\tfrac{1}{2}\langle J_z^2\rangle - \tfrac{5}{8}) . \tag{11}$$

For either mode of excitation, measurement of $(I_\sigma - I_\pi)$ vs buffer-gas pressure yields $(\gamma_2\sigma)$.

Collisional relaxation in the $^2P_{3/2}$ state also can be studied by using circularly polarized D_2 light to optically pump the $^2S_{1/2}$ ground state. $\langle S_z\rangle_g$, the electronic spin polarization of the ground state, is defined by

$$\langle S_z\rangle_g = \textstyle\sum_i n_i \langle S_z\rangle_i / \sum_i n_i , \quad \sum_i n_i = 1 \tag{12}$$

where n_i is the occupation probability of the ith ground-state sublevel, and $\langle S_z\rangle_i$ is the expectation value of the z component of electron spin for that sublevel. $\langle S_z\rangle_g$ is dependent on the degree of relaxation that occurs during the lifetime of an atom pumped to the $^2P_{3/2}$ excited state. At low buffer-gas pressures, $\langle S_z\rangle_g$ is positive, while at buffer pressures of several Torr, $\langle S_z\rangle_g$ is negative. By measuring the pressure at which $\langle S_z\rangle_g = 0$, the cross section for collisional relaxation of $\langle J_z\rangle$ in the $^2P_{3/2}$ state can be determined.[10,15-21]

For σ^+ D_2 pumping of a nuclear-spin-zero atom, the excitation rates out of the ground-state sublevels are $3An(+\frac{1}{2})$ and $An(-\frac{1}{2})$, where A is a constant proportional to the incident light intensity. The rate of change of $\langle S_z\rangle_g$ then can be written

$$\frac{d\langle S_z\rangle_g}{dt} = -2A\langle S_z\rangle_g - \tfrac{1}{2}A + \frac{N\langle J_z\rangle}{3\tau} . \tag{13}$$

In equilibrium we find

$$\langle J_z\rangle = (2\langle S_z\rangle_g + \tfrac{5}{4})(1+\Gamma_1\tau)^{-1}(1+\langle S_z\rangle_g)^{-1} , \tag{14}$$

$$N = 2A\tau(1+\langle S_z\rangle_g) , \tag{15}$$

and

$$\langle S_z\rangle_g = (2-3\Gamma_1\tau)(4+12\Gamma_1\tau)^{-1} . \tag{16}$$

Measurement of p_z, the buffer-gas pressure at which $\langle S_z\rangle_g = 0$, thus yields unique determination of $(\gamma_1\sigma)$ through the relation

$$\tfrac{2}{3} = \tau\Gamma_1 = \tau n_0 (\gamma_1\sigma) v_{\mathrm{rel}} p_z/p_0 . \tag{17}$$

EFFECT OF NUCLEAR SPIN

All real alkali-metal atoms have nonzero nuclear spin. In weak magnetic fields, the nuclear spin $\vec{I}$ and the electronic angular momentum $\vec{J}$ couple through the hyperfine interaction to form the total atomic angular momentum $\vec{F}$. The existence of the hyperfine interaction adds complexity both to the relaxation process itself and to its experimental investigation. Omont, for example, has shown that collisional relaxation in the presence of hyperfine structure is governed by $(2J+1)(2I+1)$ decay constants.[3] It is desirable to separate the effects of the collisional interaction itself, which acts primarily on the electronic system, from effects caused by the hyperfine interaction of the electronic system with the nucleus. The goal, in essence, is to obtain measurements of $(\gamma_x\sigma)$ that are independent of nuclear spin, that is, to obtain the $(\gamma_x\sigma)$ for an $I=0$ atom from measurements on an atom with $I\neq 0$.

The nuclear-decoupling approximation affords great simplification of the problem of collisional relaxation in the presence of hyperfine structure. Detailed discussions can be found in Refs. 4, 10, and 22–25. The main point is that if the duration of the collision is much shorter than the hyperfine period, the nuclear and electronic systems are decoupled, and the nucleus remains "stationary"

during the collision. In the time between collisions, the hyperfine interaction acts to reorient the nucleus. If relative probabilities for collisional transitions $|J,m_J\rangle \rightarrow |J,m_J'\rangle$ are known, application of the nuclear-decoupling approximation allows calculation of the relative collisional-transition probabilities $|F,m_F\rangle \rightarrow |F',m_F'\rangle$. This approximation reduces the number of "free" parameters to the $(2J+1)$ decay constants of the nuclear-spin-zero atom.

Previous calculations of the probabilities $|F,m_F\rangle \rightarrow |F',m_F'\rangle$ have been made *assuming* various $|J,m_J\rangle \rightarrow |J',m_J'\rangle$ models, permitting the extraction of cross sections for electronic collisional interactions, independent of nuclear spin, from experimental data. Assumption of a model for the electronic interaction clearly is an undesirable approach if the experimental data are to be used to gain detailed information on the nature of the interaction itself. There thus has been an important contrast in work on atoms with and without hyperfine structure. In the absence of hyperfine structure, decay rates Γ_1 and Γ_2 have been measured, and the corresponding cross sections $(\gamma_1\sigma)$ and $(\gamma_2\sigma)$ extracted, independent of the model.[3,25,26] The ratio γ_1/γ_2 is of value in the determination of the correct form of the interaction. In the presence of hyperfine structure, the analysis of experimental data has required the assumption of some model for the collisional interaction, thereby arbitrarily fixing the ratio γ_1/γ_2. All that can be determined in that case is the strength of the interaction as measured by the magnitude of the cross section σ, with this determination being model dependent.

An important exception to the above discussion exists in the work of Faroux.[27] Faroux has shown that certain linear combinations of the parameters $\langle \vec{J}\cdot\vec{I}\rangle$ exist which have simple relaxation characteristics, and which involve only $(\gamma_1\sigma)$ or $(\gamma_2\sigma)$. The expectation value $\langle \vec{J}\cdot\vec{I}\rangle$ within a hyperfine (F) state is essentially the total population of that F state, summed over all m_F sublevels. By making selective excitation of hyperfine states and monitoring collisionally induced transitions to other F states, Faroux was able to determine $\gamma_1\sigma$ and $\gamma_2\sigma$ for collisional relaxation in the $^3P_1$ state of Hg^{201}, in the presence of hyperfine structure. Such an approach unfortunately is not applicable to the alkali metals. Whereas the hyperfine splittings in Hg^{201} are of the order of 10^4 MHz, easily resolvable by standard optical techniques, hyperfine splittings in the $^2P_{3/2}$ states of the alkali metals are of the order of the Doppler width or less, ruling out selective excitation and monitoring of particular hyperfine states. It appears that if we are to work at low magnetic fields, and restrict ourselves to depolarization experiments, we must use classic experiments, which are easy to perform but difficult to analyze.

It is easy to show by explicit calculation that even in the presence of the hyperfine interaction, $(I_{\sigma^+}-I_{\sigma^-})(\parallel H_0)$ still monitors $\langle J_z\rangle$, and $(I_\sigma - I_\pi)(\perp H_0)$ still monitors $\langle J_z^2\rangle$:

$$(I_\sigma - I_{\sigma^-}) = \tfrac{1}{3}\langle J_z\rangle\ , \tag{18a}$$

$$(I_\sigma - I_\pi) = (\tfrac{1}{2}\langle J_z^2\rangle - \tfrac{5}{8}\ , \tag{18b}$$

where $\langle J_z\rangle$ and $\langle J_z^2\rangle$ remain defined by Eqs. (8a) and (8b) with the summation now being made over all (F,m_F) sublevels. We have yet to show how the hyperfine interaction affects the relaxation of $\langle J_z\rangle$ and of $\langle J_z^2\rangle$, and how measurement of this relaxation can be related to the $(\gamma_1\sigma)$ and $(\gamma_2\sigma)$ of the nuclear-spin-zero atom. We have attacked this problem by generalizing the calculations of Ref. 1, assuming that the nuclear decoupling approximation is valid, but making no particular assumption of model for the electronic collisional interaction. Similar calculations have been performed recently by Okunevitch and Perel.[11] The transition-probability matrix that results from this calculation is presented in Appendix A for the case of $I=\frac{3}{2}$. Similar matrices for $I=\frac{5}{2}$ and $\frac{7}{2}$ are published elsewhere.[28] These matrices, together with the transformation equations [Eqs. (6a)–(6d)], permit the calculation of the decay of expectation values of angular momentum operators in terms of γ_1, γ_2, γ_3, and σ of the nuclear-spin-zero atom.

It would be quite surprising if the generalized equations describing the relaxation of $\langle J_z\rangle$ and $\langle J_z^2\rangle$ in a $J=\frac{3}{2}$ state would continue to yield simple exponential relaxation in the presence of hyperfine structure. We are already familiar with the analogous effect of nuclear spin on collisional relaxation in the $^2S_{1/2}$ ground state.[5–9] Only in special cases is the relaxation of $\langle S_z\rangle$ simple exponential: In general it involves the sum of two exponentials. Nevertheless, for $I=\frac{3}{2}$ and $J=\frac{3}{2}$, the relaxation of $\langle J_z\rangle$ does in fact remain simple exponential, with the decay rate related to the cross section for the destruction of orientation in an $I=0$ atom $\gamma_1\sigma$ as

$$\frac{d\langle J_z\rangle}{dt} = -\tfrac{1}{2}(\gamma_1\sigma)nv_{\mathrm{rel}}\langle J_z\rangle\ . \tag{19}$$

Measurement of $[I_{\sigma^+}-I_{\sigma^-}]$ vs buffer-gas pressure in this case thus yields $\frac{1}{2}(\gamma_1\sigma)$. The only effect of hyperfine structure is to decrease the depolarization rate, compared to an $I=0$ atom, by a factor of 2. This is a very special case, which does *not* obtain for atoms of $I\neq\frac{3}{2}$. The relaxation of $\langle J_z^2\rangle$, even for $I=\frac{3}{2}$, is *not* simple exponential, and is not dependent on γ_2 alone: Its relaxation involves all three γ_x.

The D_2 optical-pumping technique is similarly complicated when applied to an alkali-metal atom

possessing hyperfine structure. In the calculation for an $I=0$ atom in which we related $(\gamma_1\,\sigma)$ to p_e [Eqs. (12)–(17)], we were able to express both the depopulation rate of $\langle S_z\rangle_g$ in the ground state and the excitation rate of $\langle J_z\rangle$ in the excited state in terms of $\langle S_z\rangle_g$. We do not obtain such a simple relation for an alkali-metal atom of $I\neq 0$. For $I=\frac{3}{2}$ the rate of change of $\langle S_z\rangle_g$ due to excitation out of the ground state is

$$\frac{d\langle S_z\rangle_g}{dt} = -4A(\langle S_z\rangle_g + \langle S_z^2\rangle_g)\ . \tag{20}$$

The rate of excitation of $\langle J_z\rangle$ in the excited state cannot be expressed in terms of $\langle S_z\rangle_g$ and $\langle S_z^2\rangle_g$. This fact makes it necessary to solve the problem in terms of individual ground-state sublevel populations rather than in terms of $\langle S_z\rangle_g$. Such a complication requires analytic inversion of the generalized 16×16 excited-state relaxation matrix, a task beyond the scope of the present, or any contemplated, work. Still, we can show that for $I=\frac{3}{2}$ the D_2 optical-pumping signal is dependent only on $(\gamma_1\,\sigma)$. For $I=\frac{3}{2}$, the repopulation rate of $\langle S_z\rangle$ due to decay from the excited state is given by

$$\frac{d\langle S_z\rangle_g}{dt} = N(3\tau)^{-1}(\langle J_z\rangle_3 + \tfrac{1}{4}\langle J_z\rangle_2 + \tfrac{1}{4}\langle J_z\rangle_1)\ . \tag{21}$$

$\langle J_z\rangle_F$ is the expectation value of J_z within the F hyperfine state. The three expectation values of $\langle J_z\rangle_F$ above can be shown to decay with the same rate, which is proportional to $(\gamma_1\sigma)$. The passage of $\langle S_z\rangle$ through zero at p_e is thus dependent on $(\gamma_1\,\sigma)$ only. However, without the explicit solution referred to above, we cannot determine the value of the numerical factor analogous to the $\frac{2}{3}$ in Eq. (17) that would allow us to extract a model independent $(\gamma_1\sigma)$. For atoms of $I\neq\frac{3}{2}$, the other γ_x also influences the passage of $\langle S_z\rangle_g$ through zero.

We have seen that the three common methods of measuring the relaxation of $\langle J_z\rangle$ and $\langle J_z^2\rangle$ continue to monitor the relaxation of these parameters in the presence of hyperfine structure. The hyperfine interaction, however, alters the relaxation process: In general it mixes all γ_x into the relaxation of any one observable. Although the three experiments described above cannot, alone, be used to determine all k_α and σ, still they can be used to obtain necessary, but not sufficient, conditions for the validity of a particular collisional model. The assumption of a particular model fixes the values of all elements of the appropriate relaxation matrix. The only remaining unknown is the cross section σ. All depolarization experiments thus can be parametrized in terms of α, the ratio of collision rate to rate of spontaneous decay:

$$\alpha = \tau n\sigma v_{\rm rel}\ . \tag{22}$$

The recent calculation of Okunevitch and Perel,[11] which is based on the van der Waals interaction, serves an example. Okunevitch and Perel have calculated the following relative probabilities: $K_0=0.22$, $K_1=0.30$, $K_2=0.27$, $K_3=0.18$, $K_A=0.25$, and $K_B=0.21$. Inserting these values into the appropriate relaxation matrices, and performing the necessary computer calculations, we find the following values for α_0, the value of α for which $\langle S_z\rangle_g$ passes through zero for D_2 optical pumping: $\alpha_0(I=\frac{3}{2})=1.97$, $\alpha_0(I=\frac{5}{2})=2.07$, and $\alpha_0=(I=\frac{7}{2})=1.97$, We also can calculate the dependence of any depolarization experiment on α. As an example, we show in Fig. 1 the dependence of $(I_\sigma-I_\pi)/(I_\sigma+I_\pi)$ on α for $I=\frac{3}{2}$, $\frac{5}{2}$ and $\frac{7}{2}$. Similar calculations can be performed for any other observable. Experimental measurements, fitted to the theoretical predictions, scale α to the actual buffer-gas pressure, yielding the value of σ. All experiments performed on a given atom must yield the same value of σ. If they do not, the model for the collisional interaction is incorrect.

We stress one final point. In using the nuclear-decoupling approximation, we have assumed that there is sufficient time between collisions for the hyperfine interaction to reorient the nuclear spin. The characteristic times involved are $\tau_{\rm hf}$, the inverse of the energy difference between adjacent hyperfine states, and τ_c, the mean time between collisions. If τ_c is considerably shorter than $\tau_{\rm hf}$, the nucleus experiences only random torques of short duration, which average to zero. We expect nuclear decoupling to be a reasonable approximation as long as $\tau_c > \frac{1}{2}\tau_{\rm hf}$. The smallest hyperfine intervals in the five $^2P_{3/2}$ states of ^{85}Rb and ^{87}Rb are about 50 MHz. Taking the lifetime of the $^2P_{3/2}$ state to be 2.7×10^{-8} sec,[29] we find, for the smallest

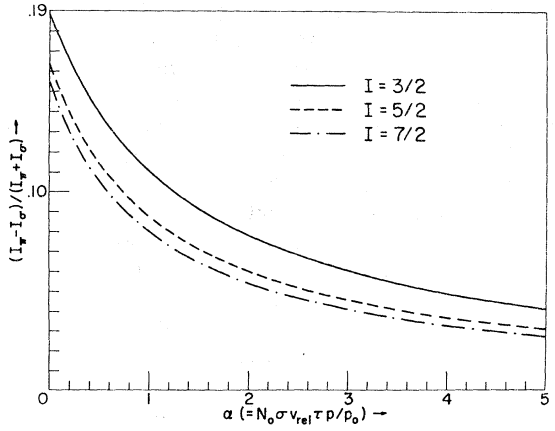

FIG. 1. Calculated dependence of the observable $(I_\pi-I_\sigma)/(I_\pi+I_\sigma)$ on collision rate for a $^2P_{3/2}$ state coupled to nuclear spins of $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$. A van der Waals model of the collisional interaction has been assumed.

TABLE IV. Values of α_0 ($=\tau n \sigma v_{\rm rel}$) at which $\langle S_z \rangle = 0$ in D_2 optical pumping of ^{85}Rb and ^{87}Rb, based on several collisional models.

Model	$\alpha_0(^{85}\mathrm{Rb})(I=\frac{5}{2})$	$\alpha_0(^{87}\mathrm{Rb})(I=\frac{3}{2})$	$\alpha_0(^{87}\mathrm{Rb})/\alpha(^{85}\mathrm{Rb})$
Δm_J random (Ref. 10)	1.90	1.80	0.95
$m_J \not\to -m_J$ (Refs. 30 and 31)	2.89	2.77	0.95
van der Waals (Ref. 11)	2.07	1.97	0.95

hyperfine interval, that $\tau_c \approx \tau_{\rm hf}$, for $\alpha = 2$, the approximate collision number at which we expect $\langle S_z \rangle$ in D_2 pumping to pass through zero. At buffer pressures more than a few Torr, that is, beyond the $\langle S_z \rangle = 0$ point, the nuclear-decoupling approximation should break down.

EXPERIMENTAL TEST OF NUCLEAR-DECOUPLING APPROXIMATION

We have used D_2 optical pumping of ^{85}Rb ($I=\frac{5}{2}$) and ^{87}Rb ($I=\frac{3}{2}$) to test the validity of the nuclear-decoupling approximation in collisional relaxation. Although the nuclear spin and hyperfine structure are different in the two isotopes, the electronic collisional interaction should be the same. We therefore should be able to use the nuclear-decoupling approximation and the calculations referred to above to predict the ratios of pressure at which $\langle S_z \rangle$ should pass through zero for ^{85}Rb and ^{87}Rb in any particular buffer gas. We cannot make these calculations in complete generality for the reasons stated in the preceding section. We have made calculations, however, in the three special cases of random reorientation of J,[1] random reorientation of J restricted by the selection rule $m_J \not\to -m_J$,[30,31] and a model based on the van der Waals interaction.[11] The results are displayed in Table IV. Different models yield different values for α, a fact already well known. However, the ratio $\alpha_0(^{87}\mathrm{Rb})/\alpha_0(^{85}\mathrm{Rb})$ turns out to be approximately 0.95 in all cases, and hence is essentially independent of the model assumed for the electronic relaxation. Since the average relative velocities of ^{85}Rb and ^{87}Rb with any particular buffer gas differ by considerably less than 1%, we draw the conclusion that if the nuclear decoupling approximation is valid, D_2 optical-pumping experiments on ^{85}Rb and ^{87}Rb for a particular buffer gas should yield a ratio of $p_z(^{87}\mathrm{Rb})/p_z(^{85}\mathrm{Rb})$ approximately equal to 0.95, independent of the exact form of the electronic collisional interaction. An experiment on the D_2 optical pumping of the Rb isotopes already has been performed by Zhitnikov, Kuleshov, and Okunevitch.[32] Their results yield values of $p_z(^{87}\mathrm{Rb})/p_z(^{85}\mathrm{Rb})$ that range from 1.29 to 1.20 and differ by as much as 35% from our predictions, throwing the validity of the nuclear decoupling approximation into question. We have measured p_z for ^{85}Rb and ^{87}Rb in He, Ne, and Ar buffer gases, using the technique to be described below. In contrast to Zhitnikov *et al.*, we find good agreement with the nuclear-decoupling approximation.

Our D_2 optical-pumping system consisted of an isotopic light source, customary lenses, polarizers, and filters, a cylindrical wax-coated cell connected to a gas handling manifold, and a photodetector. The optical-pumping cell was a Pyrex cylinder 7.6 cm in diameter, 6.8 cm long, coated with dotriacontane, and contained natural Rb (72% ^{85}Rb, 28% ^{87}Rb) in a separate reservoir. Natural Rb rather than a separated isotope was used primarily for an economic reason; wax-coated cells tend to absorb alkali-metal vapors.[33] We found that the most reproducible results were obtained if several hundred mg of the metal was available in the sidearm. A cost of more than one dollar per mg thus militates against the use of separated isotopes. With natural Rb in the cell, it is essential that the presence of one isotope not influence the optical pumping of the other. In particular, all types of spin exchange, ^{85}Rb-^{85}Rb, ^{87}Rb-^{87}Rb, ^{85}Rb-^{87}Rb, must be avoided. For this reason we enclosed the cell in Styrofoam insulation through which we passed cooled N_2 gas. The light path was kept clear and condensation free by using evacuated cylinders as windows. At the operating temperature of 17 °C, the vapor pressure of Rb is 1×10^{-7} Torr,[34] indicating a characteristic time for spin exchange of more than 1 sec. Because of combination of the alkali-metal with the wax surface, the actual number density of Rb atoms actually present in the cell may have been less than that indicated by the vapor pressure. The measured relaxation time of the evacuated cell was about 410 msec for ^{85}Rb.

The optical-pumping cell was situated in a magnetic field of 0.595 G. At this field, the Zeeman resonances of the two Rb isotopes are well enough separated (277 and 417 kHz) to permit saturation of the ground-state resonance of one isotope without affecting the spin polarization of the other. We observed transient pumping signals by monitoring the intensity of the D_2 pumping beam as rf at the resonant frequency was pulsed on and off.[35] The optical-pumping experiment on each isotope was performed using the corresponding isotopic lamp.[36,37]

The signal of interest for the present work is

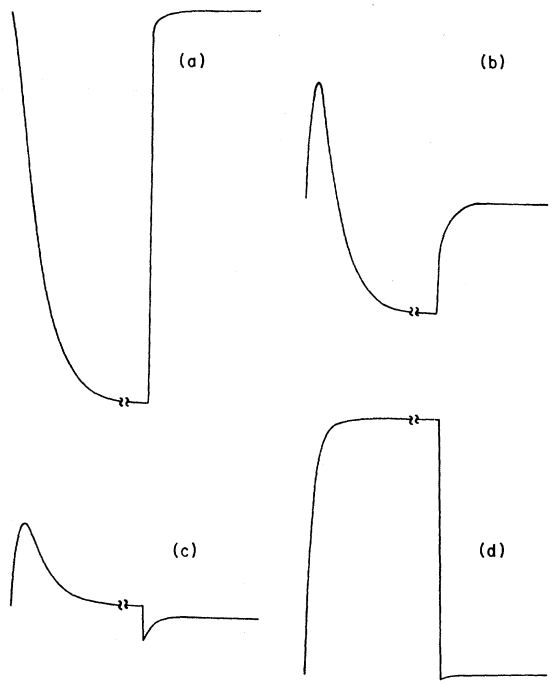

FIG. 2. Sample D_2 optical-pumping transient signals for ^{85}Rb as a function of He buffer-gas pressure. (a) 0.51 Torr, (b) 1.25 Torr, (c) 1.50 Torr, (d) 5.01 Torr. P_z the pressure at which $\langle S_z\rangle$ passes through zero was found to be 1.52 Torr. The rf is pulsed off at the left of the transient, and pulsed on at the midpoint.

not the shape of the transient signal, but rather the difference in equilibrium values for the light absorption with rf on ($\langle S_z\rangle = 0$) and with rf off ($\langle S_z\rangle = \langle S_z\rangle_{eq}$). At the buffer pressure p_z, $\langle S_z\rangle$ is equal to zero both at the start of optical pumping ($t = 0$), and in the optically pumped equilibrium state ($t = \infty$), even though the actual distributions of population throughout the Zeeman sublevels are quite different in the two cases. Thus, although the initial and final levels of the transient signal must be equal at p_z, the s ignal itself does not vanish. Sample transient signals for the D_2 optical pumping of Rb at buffer-gas pressures both above and below p_z are shown in Fig. 2.

Our detection system included a biased Schottky Barrier photodiode, a PAR113 dc preamplifier, and a Hewlett-Packard signal averager. The sweep of the signal averager was triggered synchronously with a 0.495-Hz square wave modulation of the rf power. Generally 1024 sweeps of the transient signal were made at pressures near p_z. At least three separate determinations of p_z were made for each isotope in each buffer gas. Our measured values of p_z for ^{85}Rb and ^{87}Rb in He, Ne and Ar are given in Table V.

A probable source of the discrepancy of the previously reported results of Zhitnikov *et al.* with both our theoretical predictions and experimental measurements lies in their use of unsaturated, unresolved, overlapping Zeeman resonance signals to monitor the ground-state spin polarization, a method of questionable validity.[38] Their definition of p_z as the pressure at which derivative signals in the upper and lower hyperfine levels are of the same strength is arbitrary. In contrast to the results of Zhitnikov *et al.*, we find consistency with the nuclear-decoupling approximation to within 5%.

After completion of this work, we learned of a more recent experiment of Zhitnikov, Kuleshov, Okunevitch and Sevast'yanov,[12] in which D_2 optical pumping has been used to measure $\langle S_z\rangle_z$ passing through zero for the upper hyperfine level alone. These measurements yield ratios of $\alpha'(^{87}\mathrm{Rb})/\alpha'(^{85}\mathrm{Rb})$ equal to 0.70 for He, 0.63 for Ne, and 0.64 for Ar, where $\alpha'(\mathrm{Rb})$ is the collisional number at which $\langle S_z\rangle$ is equal to zero in the upper hyperfine level. The new experiment is free of the difficulties mentioned in the preceding paragraph. We have performed the relevant calculations to find predicted values at $\alpha'(^{87}\mathrm{Rb})/\alpha'(^{85}\mathrm{Rb})$ for the three models that we have discussed, and find a value of 0.64 in all cases. The new results of Zhitnikov *et al.* for Ne and Ar thus also are in excellent agreement with the nuclear-decoupling approximation, *independent of the model of electronic relaxation.*

The assumption of a model describing the electronic relaxation allows us to extract relaxation cross sections from the data in Table V. Taking the van der Waals model of Okunevitch and Perel,[11] we find the cross-section values listed in Table VI. The model of J random reorientation would yield values about 8% smaller.

ACKNOWLEDGMENTS

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APPENDIX A: GENERALIZED RELAXATION MATRICES FOR $j=\frac{3}{2}$, $I=\frac{3}{2},\frac{5}{2},\frac{7}{2}$

In Table VII we list the generalized relaxation

TABLE V. Measured values of p_z, the buffer-gas pressure at which $\langle S_z\rangle = 0$ in D_2 optical pumping for ^{85}Rb and ^{87}Rb in He, Ne, and Ar. The ratio $p_z(^{87}\mathrm{Rb})/p_z(^{85}\mathrm{Rb})$ should be 0.95, according to the nuclear-decoupling approximation.

Buffer gas	$p_z(^{85}\mathrm{Rb})$ (Torr)	$p_z(^{87}\mathrm{Rb})$ (Torr)	$\frac{p_z(^{87}\mathrm{Rb})}{p_z(^{85}\mathrm{Rb})}$
He	1.52 ± 0.04	1.53 ± 0.04	1.00
Ne	3.44 ± 0.07	3.47 ± 0.05	1.01
Ar	1.99 ± 0.05	2.06 ± 0.06	1.03

TABLE VI. Cross sections for collisional relaxation of ^{85}Rb and ^{87}Rb in He, Ne, and Ar, evaluated from the present experimental data, subject to the assumption of the van der Waals model for the electric collisional interaction (Ref. 11). All cross sections are in units of 10^{-14} cm^2. The experimental uncertainties are estimated to be less than 10%.

Buffer gas \ Alkali metal	^{85}Rb	^{87}Rb
He	1.19	1.13
Ne	1.09	1.03
Ar	2.44	2.26

matrix $M(J,I)$ for $J=\frac{3}{2}$ and $I=\frac{3}{2}$, which, subject to the nuclear-decoupling approximation, connects (F,m_F) sublevels in a single collision. The k's refer to relative transition probabilities for a nuclear-spin-zero atom (Table I). For convenience of display, all elements have been multiplied by 200. Matrices for $J=\frac{3}{2}$ and $I=\frac{5}{2}$ and $\frac{7}{2}$ also have been calculated, and are available elsewhere.[28]

The relaxation matrix M^* summed over all collisions is given by[18]

$$M^* = (1+\alpha)^{-1}[E - \alpha M(\alpha+1)^{-1}], \qquad \text{(A1)}$$

where α is defined by Eq. (18) and E is the unit matrix.

APPENDIX B: TRANSITION PROBABILITIES BETWEEN m_J STATES

Equations (6a)–(6d) and (7) yield some informative restrictions on transition probabilities between m_J states.

(a) $\vec{J}$ reorientation restricted by a selection rule[10] $\Delta m_J = 0, \pm 2$ is not a physically possible model. Such a selection rule requires $k_0 = k_1 = k_2 = 0$, which, through Eq. (7), requires that k_2 also be zero: No relaxation can occur, a conclusion clearly contradicted by the large cross sections for relaxation in the $^2P_{3/2}$ state that have been measured.

(b) Reorientation of J restricted by the selection rule[30,31] $m_J \neq -m_J$ ($k_0 = k_3 = 0$) is not ruled out by Eqs. (6). If such a rule were rigorously valid, it would imply $\gamma_2 = 2\gamma_1$. It is interesting to note that Gallagher reports almost exactly this result in the $^2P_{3/2}$ state of Rb.[31] It is not clear, however, that Gallagher's Hanle-effect measurements in the presence of the hyperfine interaction actually yield $(\gamma_1\sigma)$ and $(\gamma_2\sigma)$.[39]

(c) γ_3 is measurable only if fluorescence from different m_J sublevels can be resolved in energy: γ_3 is not measurable at low magnetic fields. Nevertheless, Eqs. (6a)–(6d) provide upper and lower bounds on γ_3 in terms of γ_1 and γ_2, through the trivial assumption that all relaxation rates must be greater than or equal to zero. We obtain the

TABLE VII. Generalized transition probabilities for collisional relaxation among the (F, m_F) sublevels of a $J=\frac{3}{2}$ state ($I=\frac{3}{2}$). The k's are defined in Table I.

	$\|3,3\rangle$	$\|3,2\rangle$	$\|3,1\rangle$	$\|3,0\rangle$	$\|3,-1\rangle$	$\|3,-2\rangle$	$\|3,-3\rangle$	$\|2,2\rangle$	$\|2,1\rangle$	$\|2,0\rangle$	$\|2,-1\rangle$	$\|2,-2\rangle$	$\|1,1\rangle$	$\|1,0\rangle$	$\|1,-1\rangle$	$\|0,0\rangle$
$\|3,3\rangle$	$200K_A$	$100K_1$	$40K_2$	$10K_3$	0	0	0	$100K_1$	$100K_2$	$50K_3$	0	0	$60K_2$	$90K_3$	0	$50K_3$
$\|3,2\rangle$	$100K_1$	$50K_A+50K_B$	$20K_0+60K_1$	$50K_2$	$20K_3$	0	0	$50K_A+50K_B$	$50K_0$	$50K_2$	$50K_3$	0	$30K_0+40K_1$	$50K_2$	$30K_3$	$50K_2$
$\|3,1\rangle$	$40K_2$	$20K_0+60K_1$	$8K_0+80K_B$	$54K_0+20K_1$	$48K_2$	$20K_3$	0	$20K_0+60K_1$	$20K_A+20K_B$	$30K_0+20K_1$	$60K_2$	$20K_3$	$12K_A+60K_1$	$6K_0+20K_1$	$52K_2$	$30K_0+20K_1$
$\|3,0\rangle$	$10K_3$	$50K_2$	$54K_0+20K_1$	K_A+81K_B	$54K_0+20K$	$50K_2$	$10K_3$	$50K_2$	$50K_1$	$5K_A+45K_B$	$50K_1$	$50K_2$	$36K_0+30K_1$	$9K_A+9K_B$	$36K_0+30K_1$	$5K_A+45K_B$
$\|3,-1\rangle$	0	$20K_3$	$48K_2$	$54K_0+20K_1$	$8K_A+80K_B$	$20K_0+60K_1$	$40K_2$	$20K_3$	$60K_2$	$30K_0+20K_1$	$20K_A+20K_B$	$20K_0+60K_1$	$52K_2$	$6K_0+20K_1$	$12K_A+60K_B$	$30K_0+20K_1$
$\|3,-2\rangle$	0	0	$20K_3$	$50K_2$	$20K_0+60K_1$	$50K_A+50K_B$	$100K_1$	0	$50K_3$	$50K_2$	$50K_0$	$50K_A+50K_B$	$30K_3$	$50K_2$	$30K_0+40K_1$	$50K_2$
$\|3,-3\rangle$	0	0	0	$10K_3$	$40K_2$	$100K_1$	$200K_A$	0	0	$50K_3$	$100K_2$	$100K_1$	0	$90K_3$	$60K_2$	$50K_3$
$\|2,2\rangle$	$100K_1$	$50K_A+50K_B$	$20K_0+60K_1$	$50K_2$	$20K_3$	0	0	$50K_A+50K_B$	$50K_0$	$50K_2$	$50K_3$	0	$30K_0+40K_1$	$50K_2$	$30K_3$	$50K_2$
$\|2,1\rangle$	$100K_2$	$50K_0$	$20K_A+20K_B$	$50K_1$	$60K_2$	$50K_3$	0	$50K_0$	$50K_A+50K_B$	$50K_1$	0	$50K_1$	$30K_A+30K_B$	$50K_1$	$40K_2$	$50K_1$
$\|2,0\rangle$	$50K_3$	$50K_2$	$30K_0+20K_1$	$5K_A+45K_B$	$30K_0+20K_1$	$50K_2$	$50K_3$	$50K_2$	$50K_1$	$25K_A+25K_B$	$50K_1$	$50K_2$	$20K_0+30K_1$	$45K_A+5K_B$	$20K_0+30K_1$	$25K_A+25K_B$
$\|2,-1\rangle$	0	$50K_3$	$20K_A+20K_B$	$50K_1$	$20K_A+20K_B$	$50K_0$	$100K_2$	$50K_3$	0	$50K_1$	$50K_A+50K_B$	$50K_0$	$40K_2$	$50K_1$	$30K_A+30K_B$	$50K_1$
$\|2,-2\rangle$	0	0	$20K_0+60K_1$	$50K_2$	$20K_0+60K_1$	$30K_A+30K_B$	$100K_1$	0	$50K_3$	$50K_2$	$50K_0$	$50K_A+50K_B$	$30K_3$	$50K_2$	$30K_0+40K_1$	$50K_2$
$\|1,1\rangle$	$60K_2$	$30K_0+40K_1$	$12K_A+60K_B$	$36K_0+30K_1$	$52K_2$	$30K_3$	0	$30K_0+40K_1$	$30K_A+30K_B$	$20K_0+30K_1$	$40K_2$	$30K_3$	$18K_A+50K_B$	$4K_0+30K_1$	$48K_2$	$20K_0+30K_1$
$\|1,0\rangle$	$90K_3$	$50K_2$	$6K_0+20K_1$	$9K_A+9K_B$	$6K_0+20K_1$	$50K_2$	$90K_3$	$50K_2$	$50K_1$	$45K_A+5K_B$	$50K_1$	$50K_2$	$4K_0+30K_1$	$81K_A+K_B$	$4K_0+30K_1$	$45K_A+5K_B$
$\|1,-1\rangle$	0	$30K_3$	$52K_2$	$36K_0+30K_1$	$12K_A+60K_B$	$30K_0+40K_1$	$60K_2$	$30K_3$	$40K_2$	$20K_0+30K_1$	$30K_A+30K_B$	$30K_0+40K_1$	$48K_2$	$4K_0+30K_1$	$18K_A+50K_B$	$20K_0+30K_1$
$\|0,0\rangle$	$50K_3$	$50K_2$	$30K_0+20K_1$	$5K_A+45K_B$	$30K_0+20K_1$	$50K_2$	$50K_3$	$50K_2$	$50K_1$	$25K_A+25K_B$	$50K_1$	$50K_2$	$20K_0+30K_1$	$45K_A+5K_B$	$20K_0+30K_1$	$25K_A+25K_B$

following inequalities:

$$\gamma_3 \geq \tfrac{1}{9}\gamma_1 + \tfrac{5}{9}\gamma_2 , \quad \text{(B1a)}$$

$$\gamma_3 \geq \gamma_1 - \tfrac{5}{3}\gamma_2 , \quad \text{(B1b)}$$

$$\gamma_3 \leq \gamma_1 + \tfrac{5}{3}\gamma_2 , \quad \text{(B1c)}$$

$$\gamma_3 \geq -9\gamma_1 + 5\gamma_2 . \quad \text{(B1d)}$$

If, for example, $\gamma_2 = 2\gamma_1$, the relations above imply that $\gamma_1 \leq \gamma_3 \leq \frac{13}{3}\gamma_1$.

(d) Although $m_J \not\leftrightarrow -m_J$ almost certainly is not strong selection rule for relaxation within the $^2P_{3/2}$ state, still in every theoretical calculation of which we are aware, transitions in which $m_J \rightarrow -m_J$ are found to be[10,11,30,31,40–43] of the lowest probability. If, accordingly, we make the assumptions $k_1 > k_3$, $k_1 > k_0$, $k_2 > k_3$, and $k_2 > k_0$, the following inequalities result:

$$\gamma_3 \geq 6\gamma_1 - 5\gamma_2 , \quad \text{(B2a)}$$

$$\gamma_3 \leq -\tfrac{2}{3}\gamma_1 + \tfrac{5}{3}\gamma_2 , \quad \text{(B2b)}$$

$$\gamma_3 \leq -\tfrac{3}{2}\gamma_1 + \tfrac{5}{2}\gamma_2 , \quad \text{(B2c)}$$

$$\gamma_3 \leq \tfrac{1}{6}\gamma_1 + \tfrac{5}{6}\gamma_2 . \quad \text{(B2d)}$$

In the example $2\gamma_1 = \gamma_2$, Eq. (B2d) would further restrict the possible range of γ_3 to $\gamma_1 \leq \gamma_3 \leq \frac{11}{6}\gamma_1$.

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‡Alfred P. Sloan Foundation Fellow.

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