Spin-axis relaxation in spin-exchange collisions of alkali-metal atoms

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We present calculations of spin-relaxation rates of alkali-metal atoms due to the spin-axis interaction acting in binary collisions between the atoms. We show that for the high-temperature conditions of interest here, the spin-relaxation rates calculated with classical-path trajectories are nearly the same as those calculated with the distorted-wave Born approximation. We compare these calculations to recent experiments that used magnetic decoupling to isolate spin relaxation due to binary collisions from that due to the formation of triplet van der Waals molecules. The values of the spin-axis coupling coefficients deduced from measurements of binary collision rates are consistent with those deduced from molecular decoupling experiments, and follow a physically plausible scaling law for the spin-axis coupling coefficients.

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I. INTRODUCTION

Spin-exchange optical pumping [1,2] of ^{3}He uses spin exchange collisions between ^{3}He atoms and optically pumped Rb atoms to produce large quantities of highly spin-polarized ^{3}He for a variety of applications, including medical imaging [3] and spin-polarized targets [4]. The efficiency of polarized ^{3}He production is determined by two fundamental rates: the Rb-He spin-exchange rate and the Rb spin-relaxation rate. The measured spin-exchange rates [5] are in fairly good agreement with theory [6]. At the high temperatures needed for the ^{3}He spin-exchange rates to exceed the wall relaxation rates, both Rb-Rb and Rb-He relaxation limit the efficiency for spin-exchange. Although it had been assumed that collisions between alkali-metal atoms rigorously conserve the spin polarization, Bhaskar *et al.* [7] discovered that rapid spin relaxation in fact occurs in high-density optically pumped Cs, with a surprisingly large inferred spin-relaxation cross section in excess of 1 Å^2. The corresponding cross section for Rb-Rb relaxation [5,8–10], while smaller, still limits the efficiency of ^{3}He production. The still smaller cross section for K-K [8,11] relaxation suggests that, if technical difficulties are surmounted, K may be the optimum partner for spin exchange with ^{3}He [5].

A few years ago, we discovered that one-half to two-thirds of alkali-alkali spin relaxation decouples in magnetic fields of a few kG [10], making an interpretation of the relaxation exclusively in terms of binary collisions implausible. Recently, careful magnetic decoupling studies in low-pressure, isotopically pure Rb and Cs samples definitively identified one source of the field-dependent relaxation as the spin-axis interaction in triplet molecules [12]. For Rb, the nuclear quadrupole interaction in singlet molecules makes a comparable contribution at low pressures. The remaining alkali-alkali relaxation mechanism at high magnetic field is then presumably from binary collisions. It is the purpose of this paper to show that the deduced values of the spin-axis interaction from binary collisions are consistent with values recently obtained from magnetic decoupling studies of triplet molecules [12]. We hypothesize a simple scaling law for the second-order spin-orbit interaction that explains the relative magnitudes of the observed spin-axis interaction strengths. Nevertheless, the measured cross sections are in every case at least a factor of 10 larger than would be expected from *ab initio* calculations [13]. Table I contains a summary of the existing data on alkali-alkali spin relaxation.

The spin-axis interaction between two alkali-metal atoms is

$$V_1 = \frac{2\lambda}{3}\mathbf{S}\cdot(3\boldsymbol{\zeta}\boldsymbol{\zeta}-\mathbf{1})\cdot\mathbf{S}. \tag{1}$$

Here the total electron spin of the valence-electron pair is $\mathbf{S}$ and $\boldsymbol{\zeta}$ is a unit vector lying along the direction of the internuclear axis. The coefficient $\lambda(R)$, a rapidly decreasing function of interatomic separation R, is currently believed to arise from both the direct spin-dipolar coupling (averaged over the electron charge distribution) and the spin-orbit interaction in second order [13]. Accumulating evidence, from both high temperature [12,14] and low-temperature experiments [14–16], suggests that the predicted spin-axis coupling (presumably arising almost entirely from second-order, spin-orbit interactions) is too small in Cs by a factor of 3 to 4, and in Rb by a factor of more than 10 [12]. Put another way, the theoretical spin-relaxation cross sections for Rb are smaller than experiment by a factor of more than 50.

This paper carefully documents how the collisional averaging of the interaction [Eq. (1)] leads to a spin-relaxation rate. The somewhat complicated averaging can be done exactly within the limitations of the classical-path approximation, so the origin of the discrepancy cannot be due to any inadequacies of the averaging. Instead, the fault must lie with the spin-dependent potential [Eq. (1)], or with the spin-independent interatomic potential which describes the classical paths, or with the neglect of some unknown collisional relaxation mechanisms other than sudden binary collisions. For very cold collisions, where not so many partial waves are involved, one could use the distorted-wave Born approximation (DWBA) [19] to account for any limitations of the classical-path method, but at the temperatures of interest

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TABLE I. Velocity-averaged spin-relaxation cross sections $\langle v\sigma\rangle/\overline{v}$ (in 10^{-18} cm^2) for alkali-metal atoms. Zero-field ''cross sections'' include contributions from both molecular formation and binary collisions. High-field cross sections are assumed to arise solely from binary collisions. The theoretical values are calculated, at a temperature of 400 K, using estimates of the spin-axis coupling strength either from a simple scaling law or from the results of *ab initio* calculations of Ref. [13]. Also shown is the ratio of experiment to *ab initio* predictions.

Atom	Ref.	Zero-field Expt.	High-field Expt.	Scaling Theory	*ab initio* Theory	Ratio
K	[8]	2.4				
K	[17]	1.0	0.62	0.044	0.067	9.2
Rb	[8]	16				
Rb	[9]	18				
Rb	[5]	9.2				
Rb	[10]	9.3	3.4	1.4	0.061	56
Rb	[18]	15	5.6	1.4	0.061	92
Cs	[7]	203				
Cs	[14]	230	110	110	11	10

here, as we will show, the results of the classical-path approximation are within a few percent of those of the DWBA.

Our recent experiments studied the spin-relaxation rates as a function of magnetic field for the three alkali-metal atoms K, Rb, and Cs, as described in Refs. [10,12,17,18]. We assume here that the remaining alkali-metal density-dependent contributions to the relaxation rate at 12 kG arise entirely from binary collisions. The data are summarized in Table I.

II. CLASSICAL PATHS

In a binary collision at temperatures of a few hundred K, many partial waves contribute to the spin-exchange relaxation, so a classical-path treatment should be adequate. Methods for averaging over all classical-path cross sections to obtain spin-relaxation rates were given in an earlier paper by Walter *et al.* [20] (referred to in the following as WHW) for collisions between alkali-metal atoms and noble-gas atoms. In particular, the anistropic magnetic hyperfine interaction between the spin of a noble-gas nucleus and the electron spin of the alkali-metal atom, Eq. (3) of WHW, has the same tensor symmetry as the spin-axis interaction [Eq. (1)], and the detailed calculations are sufficiently similar to those of WHW that we will simply summarize the results here.

As outlined by WHW and illustrated by Fig. 1 of their paper, we can assume that the orbit of the colliding pair follows a trajectory governed by the triplet potential $V_0 = V_0(R)$. The time dependence of the internuclear separation R can be found from an equation describing the conservation of energy and the angular momentum:

$$\frac{dR}{dt} = \pm w\sqrt{1-\frac{b^2}{R^2}-\frac{2V_0}{Mw^2}}. \tag{2}$$

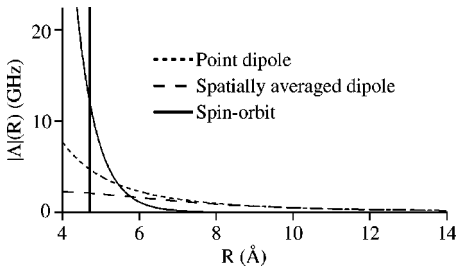

FIG. 1. *Ab initio* calculations of λ_{DD} for Rb from Ref. [13], and the modified version obtained by spatially averaging the magnetic dipole-dipole contribution as described in the text. The vertical solid line is at the classical turning point for a zero-impact parameter collision at 500-K collision energy.

Here M is the reduced mass of the pair of colliding alkali-metal atoms, and w is their relative velocity. For the high-temperature experimental conditions of interest here, possible changes in the direction of the electron spin **S** cause such small changes in the energy or angular momentum of the orbital motion that we can neglect them, and parametrize the orbital energy by the initial relative velocity w and the angular momentum by the impact parameter b.

It is convenient to let the time of closest approach of the pair be $t=0$, so the orbital angle at time t is

$$\psi = \psi(t) = \cos^{-1}[\boldsymbol{\zeta}(t)\cdot\boldsymbol{\zeta}(0)]. \tag{3}$$

The time dependence of the orbital angle can be found by a numerical quadrature of the equation for conservation of the angular momentum:

$$\frac{d\psi}{dt} = \frac{wb}{R^2}. \tag{4}$$

When averaged over a thermal distribution of trajectories, collisions in an alkali-metal vapor of atomic number density n will cause the mean longitudinal electron-spin polarization $\langle S_z\rangle$ of the atoms to relax at the rate

$$\frac{d}{dt}\langle S_z\rangle = -n\langle v\sigma\rangle\langle S_z\rangle. \tag{5}$$

The rate coefficient can be readily calculated by methods analogous to those used by WHW for relaxation due to the anisotropic magnetic dipole hyperfine interaction. The average over all angles of the collisions can be carried out analytically and we find, in analogy to Eq. (33) of WHW,

$$\langle v\sigma\rangle = \int_0^\infty dw\, p(w) w \int_0^\infty db\, b\frac{8\pi}{3}\sum_{m=-2}^{2}|\varphi_{2m}|^2. \tag{6}$$

The probability $p(w)dw$, of finding the magnitude w of the relative velocity of the colliding pair between w and $w+dw$, is

$$p(w)dw = 4\pi w^2\left(\frac{M}{2\pi k_B T}\right)^{3/2} e^{-Mw^2/2k_BT}dw, \tag{7}$$

where T is the absolute temperature and k_B is Boltzmann's constant. The tensor phases accumulated during the collision are

$$\varphi_{2m}=\frac{1}{\hbar}\int_{-\infty}^{\infty} dt\,\lambda d^2_{0m}(\psi). \tag{8}$$

Here $d^2_{0m}(\psi)$ is a Wigner d function, for example, $d^2_{00}(\psi)=(3\cos^2\psi-1)/2$ [21]. In the integrand of Eq. (8), both the spin-axis coupling coefficient λ and the orbital angle ψ are functions of time t, obtained from numerical integration of Eqs. (2) and (4). Since $\psi(-t)=-\psi(t)$, $d_{2m}(\psi)$ is an even function of t if m is even, and odd if m is odd. Also λ is an even function of t (measured from the time of closest approach), so ϕ_{2m} is identically zero if $m=\pm 1$. In practice, the rapid decrease of λ with increasing internuclear separation r means that $\phi_{2,\pm 2}$ is so small as to be negligible in practice, and so the $m=0$ contribution to Eq. (6) dominates.

III. PARTIAL WAVES

We can also calculate the spin relaxation due to the spin-axis interaction [Eq. (1)] by the DWBA, outlined by Newbury *et al.* [19], which we refer to as Newbury in this section. According to Eq. (47) of Newbury, the rate coefficient corresponding to our Eq. (6) is

$$\langle v\sigma\rangle=\int_0^{\infty} dw\,p(w)w\frac{64\pi M^2}{3\hbar^4k^6}\sum_{l=0}^{\infty}(2l+1)\lambda^2_{ll}. \tag{9}$$

For the high-temperature conditions of interest here, the three matrix elements $\lambda_{ll'}$ for spin-flip scattering from an initial wave of angular momentum l to a final wave of angular momentum $l'=l,l\pm 2$ are nearly equal. We have assumed exact equality to reduce the double sum on l and l' to a single sum on l in Eq. (9).

The classical initial relative velocity w is related to the asymptotic spatial frequency k of the scattered wave by

$$w=\frac{\hbar k}{M}. \tag{10}$$

In Eq. (9), the integral over impact parameters b, which occurs in the classical-path expression [Eq. (6)] is replaced by a sum over partial waves l. The matrix element of the spin-axis coupling coefficient between partial waves l and l' is

$$\lambda_{ll'}=k\int_0^{\infty} g_l(r)\lambda(r)g_{l'}(r)dr. \tag{11}$$

The wave functions $g_l(r)$ are solutions of the Schrödinger equation

$$\left(-\frac{d^2}{dr^2}+\frac{l(l+1)}{r^2}+\frac{2M}{\hbar^2}V_0-k^2\right)g_l=0, \tag{12}$$

with the boundary condition as $r\to 0$,

$$g_l\to 0, \tag{13}$$

and with the asymptotic boundary condition for $kr/l\to\infty$:

$$g_l\to \sin\left(kr-\frac{\pi l}{2}+\delta_l\right). \tag{14}$$

From comparison of Eq. (6) with Eq. (9), we conclude that the classical-path and partial-wave treatment will give practically the same answers if

$$\sum_{m=-2}^{2}|\varphi_{2m}|^2=\left(\frac{2\lambda_{ll}}{E}\right)^2. \tag{15}$$

In Eq. (15), we have assumed that $l=kb$. The initial relative energy of the colliding pair is

$$E=\frac{\hbar^2k^2}{2M}. \tag{16}$$

Numerical solutions to the differential equations (2) and (12) are readily obtained. We have confirmed that relation (15) is indeed true to an accuracy of about 1%, establishing the equivalence of the classical-path and partial-wave methods of calculating the spin-relaxation rate coefficients. Although the two methods give the same results, the classical-path approach is much less numerically intensive, requiring only the solution of the simple first-order differential equations (2) and (4), whereas the partial-wave analysis requires solving the second-order equation (12), with rapidly oscillating solutions. To our knowledge, this is the first quantitative comparison of partial-wave and classical-path theories for these spin-dependent cross sections.

IV. COMPARISON OF *AB INITIO* CALCULATIONS TO EXPERIMENT

In the context of ultracold collisions, Mies *et al.* [13] recently published *ab initio* calculations of λ, as the sum of two contributions:

$$\lambda=\lambda_{\mathrm{SO}}+\lambda_{\mathrm{DD}}. \tag{17}$$

For heavier alkali-metal atoms, and for small R, Mies *et al.* found that second-order spin-orbit interactions, represented in Eq. (17) by λ_{SO}, were much larger than the term λ_{DD}, which describes the direct interaction between the magnetic moments of the two valence electrons. Second-order contributions analogous to those responsible for λ_{SO} are well known from the theoretical literature on molecular spectroscopy [22,23]. Mies *et al.* parametrized their calculations of λ as follows (we have converted their results from atomic units):

$$\lambda=\frac{3g_S^2\mu_B^2}{4a_B^3}\left[Ce^{-\beta(R-R_S)}-\left(\frac{a_B}{R}\right)^3\right]. \tag{18}$$

Here a_B is the Bohr radius, μ_B is the Bohr magneton, and $g_S=2.00232$ is the electronic g-factor. The result [Eq. (18)] of the *ab initio* calculations are parametrized as follows: for Rb, $R_S=5.292$ Å, $C=0.001252$, and $\beta=1.84$ Å^{-1}; for Cs,

$R_S = 5.292$ Å, $C = 0.02249$, and $\beta = 1.568$ Å^{-1}. The first term in Eq. (18) represents λ_{SO}. The second term

$$\lambda_{DD} = -\frac{3g_S^2\mu_B^2}{4R^3} \tag{19}$$

represents the magnetic interaction of electrons, taken as point particles separated by a distance R. This is an excellent approximation at very large R, but should be modified, as we show below, at smaller values of R where the most important contributions to spin relaxation occur. This modification, however, is unlikely to have a major impact on the predictions of Ref. [13].

Table I shows cross sections calculated as described in Secs. II and III. For K, we assume only the classical spin-dipolar term, because the spin-orbit contribution estimated by scaling from Mies *et al.* [13] is negligible. As can be seen from Table I, the theoretical estimates are smaller than experiment by about a factor of 10 for Cs and K, and by a factor of almost 60 for Rb, where the *ab initio* calculations predict that λ goes to zero at $R = 5.5$ Å.

In order to describe the effects of spin relaxation in a number of experiments on Cs, where Eq. (18) predicts relaxation rates that are much too small, the National Institute of Standards and Technology group chose to multiply λ_{SO} (the computed contribution to λ from second-order spin-orbit interactions) by a constant value. This assumes that the R dependence of the calculation is correct [14,16]. Guided by new experimental data, we will discuss similar scaling arguments in Sec. VI.

V. EFFECTS OF WAVE-FUNCTION OVERLAP ON λ_{DD}

Expression (19) of λ_{DD} neglects the spatial distribution of the electron charge. In this section we present a simple estimate of the effect of the spatial distribution, and find that its neglect cannot be responsible for the discrepancy between experiment and theory.

A simple estimate of the wavefunction of the triplet state of an alkali-metal dimer is

$$|\Psi\rangle = \psi(\mathbf{r}_1,\mathbf{r}_2)|\chi\rangle = N[\varphi_A(1)\varphi_B(2) - \varphi_B(1)\varphi_A(2)]|\chi\rangle \tag{20}$$

where, for example, $\varphi_A(1)$ is a spatial orbital for electron 1 centered at nucleus A, N is a normalizing factor, and $|\chi\rangle$ is a three-component spinor representing the triplet spin state. The matrix element between a final triplet state $|\Psi_f\rangle$ and an initial triplet state $|\Psi_i\rangle$ of the electronic magnetic dipole interaction (at fixed R) is

$$\left\langle \Psi_f \left| \frac{g_S^2\mu_B^2}{r_{12}^5}\mathbf{S}_1\cdot(r_{12}^2\mathbf{1} - 3\mathbf{r}_{12}\mathbf{r}_{12})\cdot\mathbf{S}_2 \right| \Psi_i \right\rangle = \frac{4\lambda_{DD}}{3}\langle\chi_f|\mathbf{S}_1\cdot(3\boldsymbol{\zeta}\boldsymbol{\zeta} - \mathbf{1})\cdot\mathbf{S}_2|\chi_i\rangle, \tag{21}$$

where

$$\lambda_{DD} = \frac{3g_S^2\mu_B^2}{4}\int d^3r_1 d^3r_2 \frac{r_{12}^2 - 3z_{12}^2}{2r_{12}^5}|\psi(\mathbf{r}_1,\mathbf{r}_2)|^2. \tag{22}$$

We make the simplifying assumption that φ_A and φ_B can be approximated by the ground-state wave function of the valence electron of an isolated alkali-metal atom.

The results are shown in Fig. 1. The principal effect of the wave-function overlap is to reduce the value of λ_{DD} as compared to the point-dipole approximation, and to reduce the predicted *ab initio* cross section for Rb-Rb to 5.3×10^{-20} cm^2, increasing the discrepancy between experiment and theory. Use of better electron wave functions than the simple form of Eq. (20) is unlikely to change the results by the orders of magnitude needed to obtain agreement with experiment.

VI. SCALING RELATION FOR λ_{SO}

In this section we show that the spin-axis coupling coefficients deduced from high-temperature experiments on triplet molecules are consistent with a simple and plausible scaling relation that, in turn, accurately predicts the relative binary spin-relaxation cross sections. Clearly, the contribution λ_{DD} from the direct interaction of the magnetic dipole moments of the electrons is much too small to account for the observed relaxation rates in the heavier alkali-metal atoms. The *ab initio* calculation of the additional contribution λ_{SO} is also too small. In order to test the consistency of the observed molecular and binary relaxation rates, we shall assume that Mies *et al.* correctly identified the second-order spin-orbit interaction as a major contributor to λ, which implies that λ_{SO} should be proportional to the square of the $P_{1/2}$-$P_{3/2}$ fine-structure splitting $\Delta\nu$, and inversely proportional to the valence-electron binding energy E, as predicted by perturbation theory. To obtain the radial dependence of λ_{SO}, we shall make the physically plausible assumption that it scales as $|\phi(r)|^2$, the valence-electron density of an unperturbed alkali-metal atom at a distance r from the nucleus. (The radial dependence of the values of λ_{SO} calculated by Mies *et al.*, is very nearly that of $|\phi(r)|^2$.) Thus our scaling law is

$$\lambda_{SO} = \Omega\frac{(h\Delta\nu)^2}{E}|\phi(r)|^2. \tag{23}$$

The-fine structure splittings $\Delta\nu/c$ for Cs, Rb, and K are 554, 237.6, and 57.7 cm^{-1}, respectively; the binding energies E are 3.89, 4.18, and 4.34 eV.

For the required wave functions $\phi(r)$, we use the asymptotic expansion of the Coulomb wave function [25], namely,

$$\phi(r) = \frac{N}{r}\left(\frac{2r}{n^*}\right)^{n^*} e^{-r/n^*}, \tag{24}$$

where the radius r is measured in Bohr radii a_B. The effective principal quantum number n^* of the valence electron

in its ground state is related to the ionization energy E (in eV) and the Rydberg $R_\infty = 13.61$ eV by $n^* = (R_\infty/E)^{1/2}$, and the normalization factor N is given by $N = [(4\pi)^{1/2}(n^*)^{3/2}\Gamma(n^*)]^{-1}$, where $\Gamma(x)$ is the Euler gamma function.

The universal constant Ω of Eq. (23), which has units of volume, is deduced from experiments as follows. Recent experimental studies [12] of the Cs spin relaxation in triplet dimers yielded a spin-axis coupling $|\lambda^{Cs}/h| = 2.79$ GHz. This value reflects a thermal average over the rovibrational states of the triplet molecules, but for simplicity we will take it to be the value of λ/h at the Cs triplet dimer equilibrium internuclear separation of $R = 12a_B$; thus by Eqs. (17) and (19), $\lambda_{SO}^{Cs} = 2.94$ GHz. Thus, using $|\phi|^2 = 3.48 \times 10^{-6} a_B^{-3}$ at $R = 12a_B$, we obtain

$$\Omega = 2880 a_B^3, \tag{25}$$

roughly an atomic volume.

In Rb, the equilibrium internuclear separation for triplet molecules is $R = 11.5a_B$, where $|\phi|^2 = 3.23 \times 10^{-6} a_B^{-3}$, from which Eqs. (23) and (19) yield $\lambda^{Rb}/h = 294$ MHz, in close agreement with the value $|\lambda^{Rb}/h| = 290$ MHz deduced from the observation of spin relaxation due to Rb triplet dimers [12].

Using the results of Secs. II and III and the spin-axis coupling coefficients deduced from the above simple arguments, we may readily compute the spin relaxation due to the spin-axis interaction in binary collisions between alkali-metal atoms. The calculated velocity-averaged cross sections for K-K, Rb-Rb, and Cs-Cs at 400 K, using the well-known *ab initio* interatomic potentials of Krauss and Stevens [24] to compute the needed trajectories, are 4.4×10^{-20}, 1.4×10^{-18}, and 1.1×10^{-16} cm^2, respectively. As can be seen from Table I, these values agree with experiment to within a factor of 3 for rubidium and cesium. We note that for potassium it is the dipole-dipole interaction that dominates the contribution to λ, and so the larger discrepancy with experiment for K may be due to the simplified estimate [Eq. (19)] of λ_{DD} [25].

VII. CONCLUSIONS

We have shown here that the strength of the spin-axis coupling deduced from measurements of binary collisions between alkali-metal atoms is consistent with the values deduced from magnetic decoupling of relaxation due to formation of weakly bound triplet molecules. The deduced spin-axis coupling strengths are much larger than predicted by *ab initio* theory. Clearly more reliable theoretical estimates of the spin-axis coupling are needed.

Note added. Very recently Kotochigova *et al.* [26] published new calculations of the second-order spin-orbit interaction for Cs (Rb) that are roughly a factor of 2 (5) times larger than those of Mies *et al.* [13]. They have not evaluated the implications of their calculations for our high-temperature experiments.

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