Excitation transfer between the rubidium 5 ^2D fine-structure levels in collisions with ground-state rubidium atoms: Experiment and theory

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We report a study of fine-structure mixing $\mathrm{Rb}(5\ ^2D_{5/2}) \rightarrow \mathrm{Rb}(5\ ^2D_{3/2})$ in collisions with ground-state Rb atoms. In the experiment, two-photon cw laser excitation was applied to the Rb vapor cell. The measured cross section for the process was $(5.8 \pm 1.9) \times 10^{-14}\,\mathrm{cm}^2$. Theoretical calculations using nonadiabatic collision theory gave a value of $3.4 \times 10^{-14}\,\mathrm{cm}^2$.

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I. INTRODUCTION

The problem of fine-structure (FS) changing collisions

$$\mathrm{Rb}(5\ ^2D_{5/2}) + \mathrm{Rb}(5\ ^2S_{1/2}) \rightarrow \mathrm{Rb}(5\ ^2D_{3/2}) + \mathrm{Rb}(5\ ^2S_{1/2}) + \Delta E, \quad (1)$$

has attracted our attention both from an experimental and a theoretical point of view. The cross section for that process was earlier reported only in Ref. [1]. The authors used stepwise excitation, with a spectral lamp in the first step and a cw dye laser in the second step. Experimental cross sections for fine-structure (FS) mixing for the higher $\mathrm{Rb}(n\ ^2D)$ states, with n ranging from 6 to 9, were reported in the same paper, as well as in the papers [2–5] by another group. In the latter studies a pulsed two-photon dye-laser excitation was applied and the values obtained by this group are systematically higher than the respective ones of Ref. [1]. The authors of [1] attributed this discrepancy to some possible side effects which in their opinion might be associated with the pulsed laser excitation method used in [2–5]. Recently, the cross section for $n=6$ was remeasured by using pulsed two-photon excitation [6]. The obtained value turned out to be intermediate between the relevant results of the two (previously mentioned) groups. In the present work we have decided to provide a lacking counterpart to the cross section for $n=5$ of Ref. [1], by also using two-photon but cw and not pulsed excitation of Rb atoms.

The theoretical treatment of the FS-mixing collisions is reviewed by Beigman and Lebedev [7]. It should be noted, however, that the methods discussed in [7] concern states with high n values, and they are not valid for states as low as the $\mathrm{Rb}(5\ ^2D)$ state. To the best of our knowledge, the cross section for process (1) has not yet been calculated. Our theoretical calculations are based on Rb_2 molecular terms obtained in [8,9] for internuclear distances $R \geqslant 20$ a.u. by using the asymptotic approximation.

II. EXPERIMENT

A. The experimental setup and procedure

The 5 ^2P doublet in rubidium is situated nearly half-way in energy from the ground state 5 ^2S to the 5 ^2D state (Fig. 1). For this reason, two-photon excitation to 5 ^2D, and especially to the 5 $^2D_{5/2}$ component, is relatively efficient [10] even with the use of a low power cw diode laser. Recently, high precision frequency measurements of the hyperfine components of the 5 $^2S_{1/2} \rightarrow 5\ ^2D_J$ transition were performed with such excitation [11,12]. In our study of FS-changing collisions, a home-made external-cavity diode laser in the Littrow configuration was tuned to selectively populate the 5 $^2D_{5/2}$ level. The effective laser power at the required wavelength (778.1 nm) was approximately 10 mW.

The scheme of the apparatus is presented in Fig. 2. The experiment was performed with the use of a sealed-off spec-

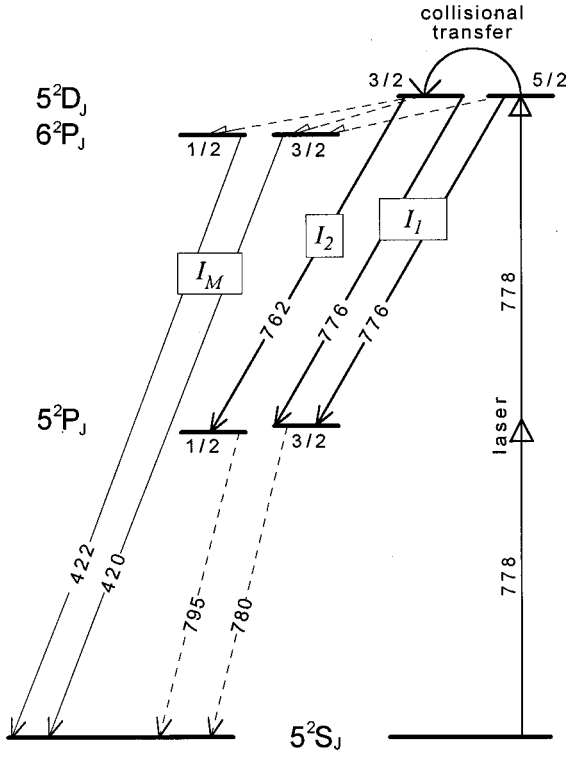

FIG. 1. The excitation and detection scheme. I_1 and I_2 are the registered fluorescence intensities to be compared, and I_M is the monitoring signal. The unobserved transitions are symbolized by dashed lines. The J values of each level are indicated.

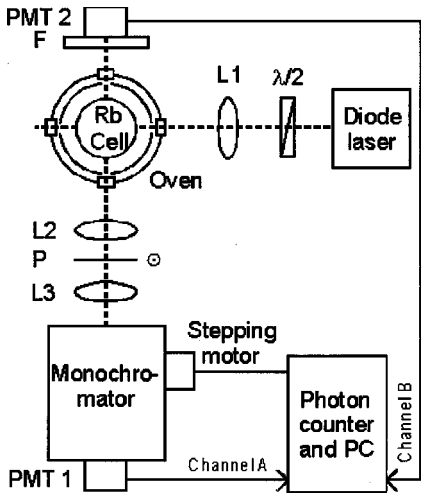

FIG. 2. A simplified schematic diagram of the experimental arrangement. PMT1 and PMT2, photomultipliers; *F*, blue filter; λ/2, half-wave plate; *P*, linear polarizer; $L1-L3$, lenses.

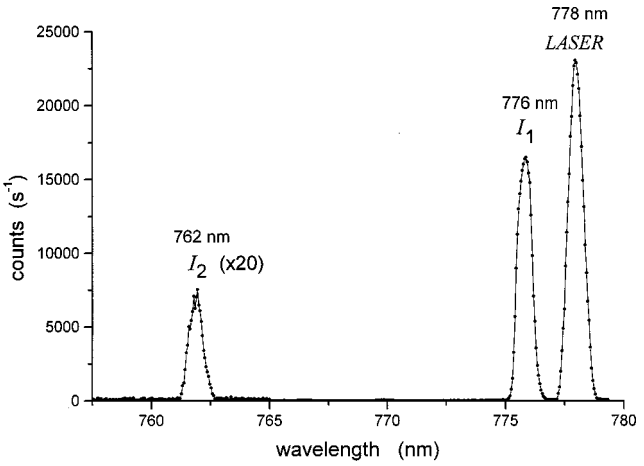

FIG. 3. An example of the registered spectra. The temperature of the cell was 403 K.

troscopic rubidium cell made of Pyrex glass. The cell was surrounded by an antimagnetic shield and placed in a double chamber oven, whose temperature was stabilized independently in its lower and upper part. The temperature of the coldest part of the cell containing a droplet of rubidium was varied in the range of 365–403 K. The temperature T of the region of excitation in the upper part was maintained 3–5 K higher, which prevented condensation of Rb on the walls. The number density of Rb atoms was from $\sim 3\times 10^{12}\,\mathrm{cm}^{-3}$ to $\sim 3\times 10^{13}\,\mathrm{cm}^{-3}$. (For the pressure-temperature relation, the Nesmeyanov formula [13] was used.)

The excitation-detection scheme provided a ''magic angle'' configuration in order to avoid polarization effects [14]. For this goal the properly oriented half-wave plate (λ/2) and the polarizer (P) were inserted across the laser beam and across the fluorescence beam, respectively. To analyze the fluorescence, a monochromator was used with a photomultiplier PMT 1 (Hamamatsu R943-02), whose output was directed to channel A of a photon counter (Stanford Research type SR400). In each PC-controlled experimental run, a spectrum of fluorescence (Fig. 3) in a range comprising peaks of the direct and sensitized fluorescence was registered at a given Rb cell temperature. The I_1 peak is the dominantly direct fluorescence, and I_2 is the sensitized one. Simultaneously, the fluorescence signal I_M at ~420 nm, corresponding to the transitions $6\,^2P_J \rightarrow 5\,^2S_{1/2}$, was selected by a blue colored glass filter and registered in channel B of SR400, with the help of a photomultiplier PMT2 (Philips PM2254B). In this way the stability of excitation was constantly monitored, since this fluorescence emerged in a cascade $5D \rightarrow 6P \rightarrow 5S$, provided the laser wavelength was in resonance with the two-photon $5S \rightarrow 5D$ transition. More details of the experiment, and in particular of the construction and performance of the diode laser, can be found in [15].

B. Rate equation and data processing

The thermally averaged effective cross section σ for a process of excitation transfer (ET) in collisions with ground-state atoms at number density n is related to the relevant transfer rate by $R_{\mathrm{ET}}=n\overline{v}^{\mathrm{rel}}\sigma$, where $\overline{v}^{\mathrm{rel}}=\sqrt{8kT/\pi\mu}$ is the mean relative velocity of the colliding partners, which are characterized by their reduced mass μ.

In the analysis of experimental data it was taken into account that, besides the collisions with the parent ground-state atoms, also collisions with some impurities present in the cell could contribute to excitation transfer from the Rb($5\,^2D_{5/2}$) state. On the basis of arguments presented in our previous paper [6], we regard helium, which can penetrate through the cell walls from the atmosphere, as a non-negligible impurity. Relevant terms were introduced in the following rate equation, by which population changes of the collisionally excited Rb($5\,^2D_{3/2}$) state were described:

$$\frac{dN_{3/2}}{dt}=N_{5/2}Nv\,\sigma_{5/2\rightarrow 3/2}+N_{5/2}N_{\mathrm{He}}v^{\mathrm{Rb\text{-}He}}\sigma^{\mathrm{Rb\text{-}He}}_{5/2\rightarrow 3/2}$$
$$-N_{3/2}\left[\frac{1}{\tau_{3/2}}+Nv(\sigma_{3/2\rightarrow 5/2}+\sigma_{3/2})\right.$$
$$\left.+N_{\mathrm{He}}v^{\mathrm{Rb\text{-}He}}(\sigma^{\mathrm{Rb\text{-}He}}_{3/2\rightarrow 5/2}+\sigma^{\mathrm{Rb\text{-}He}}_{3/2})\right]. \qquad (2)$$

Here, N, $N_{3/2}$, $N_{5/2}$, and N_{He} denote the number density of the ground-state Rb($5\,^2S_{1/2}$), Rb($5\,^2D_{3/2}$), Rb($5\,^2D_{5/2}$), and He atoms, respectively; $\tau_{3/2}$ is the natural lifetime of $5\,^2D_{3/2}$; v stands for the mean relative velocity $\overline{v}^{\mathrm{rel}}$ of two colliding Rb atoms; $\sigma_{5/2\rightarrow 3/2}$ and $\sigma_{3/2}$ are the cross sections for the processes induced by collisions with Rb atoms, respectively, for FS-mixing Rb($5\,^2D_{5/2}$)→Rb($5\,^2D_{3/2}$) and for quenching of $5\,^2D_{3/2}$, while the corresponding symbols characterizing analogous excitation processes but due to collisions of Rb with He are indexed with ''Rb-He.''

With $dN_{3/2}/dt=0$, at cw excitation, one can rearrange Eq. (2) to obtain the ratio $N_{5/2}/N_{3/2}$ as a function of $1/Nv$

$$\frac{N_{5/2}}{N_{3/2}}=\frac{\left(\frac{1}{\tau_{3/2}\sigma_{5/2\to 3/2}}+N_{\mathrm{He}}v^{\text{Rb-He}}\frac{\sigma^{\text{Rb-He}}_{3/2\to 5/2}+\sigma^{\text{Rb-He}}_{3/2}}{\sigma_{5/2\to 3/2}}\right)\frac{1}{Nv}+\frac{\sigma_{3/2\to 5/2}+\sigma_{3/2}}{\sigma_{5/2\to 3/2}}}{1+N_{\mathrm{He}}v^{\text{Rb-He}}\frac{\sigma^{\text{Rb-He}}_{5/2\to 3/2}}{\sigma_{5/2\to 3/2}}\frac{1}{Nv}}. \tag{3}$$

The formula (3) was then expressed in a simplified approximate form:

$$\frac{N_{5/2}}{N_{3/2}}=\frac{(a+1.51b)\frac{1}{Nv}+c}{1+b\frac{1}{Nv}}, \tag{4}$$

where $a=(1/\tau_{3/2})(1/\sigma_{5/2\to 3/2})$, $b=N_{\mathrm{He}}v^{\text{Rb-He}}(\sigma^{\text{Rb-He}}_{5/2\to 3/2}/\sigma_{5/2\to 3/2})$, $c=1.51+(\sigma_{3/2}/\sigma_{5/2\to 3/2})$, while $\sigma^{\text{Rb-He}}_{3/2}$ was neglected; $v^{\text{Rb-He}}$, the factor in b, which exhibits only $\sim$5% change within the temperature limits, was considered to be constant at its average value. In order to derive Eq. (4), we applied twice the detailed balancing principle: $\sigma^{\text{Rb-He}}_{5/2\to 3/2}/\sigma^{\text{Rb-He}}_{3/2\to 5/2}=\sigma_{5/2\to 3/2}/\sigma_{3/2\to 5/2}=(g_{5/2}/g_{3/2})\exp(\Delta E/kT)=1.51$, where $g_{3/2}$ and $g_{5/2}$ are the statistical weights of the FS components of the $5\ ^2D$ state, and $\Delta E=E_{5/2}-E_{3/2}=2.96\,\mathrm{cm}^{-1}$ is their splitting. We assumed that for the $5\ ^2D$ state, the ratio $\sigma^{\text{Rb-He}}_{3/2}/\sigma^{\text{Rb-He}}_{3/2\to 5/2}$ is approximately the same as the ratio of such measured cross sections available for $6\ ^2D$ from [4]. In this way we estimate that $\sigma^{\text{Rb-He}}_{3/2}$ is at most few percent of $\sigma^{\text{Rb-He}}_{3/2\to 5/2}$ and can be neglected in Eq. (4), especially since a, the value to be determined to get $\sigma_{5/2\to 3/2}$, is over one order of magnitude larger than b. If, similarly, for $\sigma_{3/2}/\sigma_{5/2\to 3/2}$ of $5\ ^2D$, the ratio of the respective values measured for $6\ ^2D$ [4] was assumed, the range 1.8–2.8 was obtained as extreme limits for c.

The measured fluorescence intensities I_1 and I_2 (Fig. 3) are related to number densities $N_{5/2}$ and $N_{3/2}$ through Einstein coefficients A_{ik} of the observed transitions. I_1 is the direct fluorescence $I_{5D_{5/2}\to 5P_{3/2}}$ with an admixture of the spectrally unresolved component $I_{5D_{3/2}\to 5P_{3/2}}$ of the sensitized fluorescence, which is small at the temperatures of the experiment:

$$\begin{aligned} I_1&=I_{5D_{5/2}\to 5P_{3/2}}+I_{5D_{3/2}\to 5P_{3/2}}\\ &=N_{5/2}A_{5D_{5/2}\to 5P_{3/2}}+N_{3/2}A_{5D_{3/2}\to 5P_{3/2}}, \end{aligned} \tag{5}$$

and I_2 is the sensitized fluorescence intensity:

$$I_2=I_{5D_{3/2}\to 5P_{1/2}}=N_{3/2}A_{5D_{3/2}\to 5P_{1/2}}. \tag{6}$$

With the numerical values of Einstein coefficients taken according to the values of oscillator strengths calculated by Hansen [16], we obtained from Eqs. (5) and (6) the following relation for I_1/I_2:

$$\frac{I_1}{I_2}=0.23+1.35\frac{N_{5/2}}{N_{3/2}}. \tag{7}$$

The formula (7) with $N_{5/2}/N_{3/2}$ expressed by Eq. (4) was fitted to the experimental dependence of I_1/I_2 versus $1/Nv$ (Fig. 4). The adjustable parameters of the fit were a and b. The value of c was usually fixed at 2.3, however if another value within the 1.8–2.8 limit was taken for c, this affected the fitted value for a by less than 2%. We note that in order to simplify Eq. (3) into Eq. (4) we use ratios of experimental cross-section values for $6\ ^2D$, which in energy is the next D state to $5\ ^2D$. We have also checked that using such ratios available for the two higher states, for $7\ ^2D$ from [2] and for $8\ ^2D$ from [3], the procedure would give a values that were at most a few percent different. The value of a, with 239 ns for lifetime $\tau_{3/2}$, taken from the calculations of Theodosiou [17], leads to the cross section $\sigma_{5/2\to 3/2}=[(5.8\pm 1.9)\times 10^{-14}]\,\mathrm{cm}^2$. In estimating the error limits, the following sources of error were accounted for: the uncertainty in Nv, in the spectral calibration, in the experimentally estimated correction for the effect of thermal escape out of the viewing region [18], as well as the statistical error of the fit and the uncertainty in the choice of c value.

It is worth mentioning that an estimate based on the value of b obtained in the fitting procedure and on helium number density in the cell, calculated by using transport rates for helium through Pyrex glass from the literature [6], gave for the ratio $\sigma^{\text{Rb-He}}_{5/2\to 3/2}/\sigma_{5/2\to 3/2}$ a value consistent with such ratios measured for the next three higher $n\ ^2D$ states [4,2,3]. This appears to support the hypothesis that helium is the non-

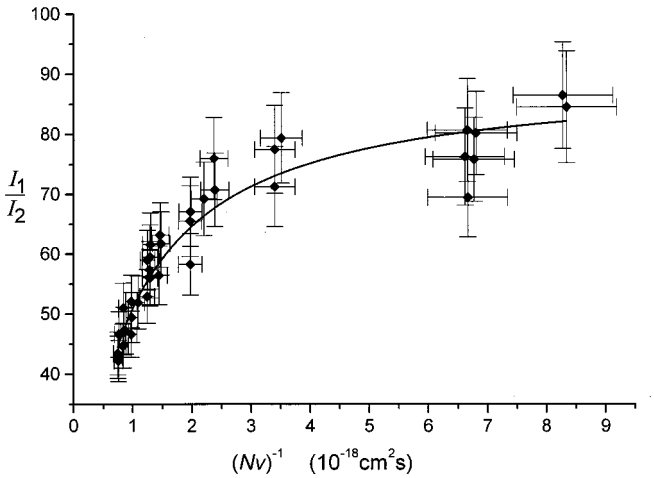

FIG. 4. Ratios of the measured fluorescence intensities vs $1/Nv$. The continuous curve is the result of fitting.

negligible impurity contributing to the collisional excitation transfer in the cell used in the experiment.

III. THEORY

Since the energy defect for FS-mixing collisions [Eq. (1)] is $\Delta E = 2.96\,\mathrm{cm}^{-1}$, the process is practically resonant. The effective cross section for this process is determined by nonadiabatic transitions between the adiabatic terms of the Rb_2 quasimolecule which correlate with the $5\,^2D_{5/2} + 5\,^2S_{1/2}$ and $5\,^2D_{3/2} + 5\,^2S_{1/2}$ states of separated atoms. The nearly resonant character of the process and the large experimental value of the cross section indicate that the main contribution to the FS mixing is due to nonadiabatic transitions at large internuclear distances $R \geqslant 30$ a.u. The Rb_2 terms for large R have previously been calculated in [8,9,19] by applying the simple and efficient asymptotic method [20]. These terms [8,9] were used in the present work to calculate the $5\,^2D$ state FS-mixing cross section.

In the Rb_2 quasimolecule composed of an atom in the $5\,^2D$ state and an atom in the $5\,^2S$ state, there are two types of term splitting at large internuclear distances. One of them is determined by the spin-orbit interaction of the atomic valence electron and it is independent of the distance between the atoms. The other is related to electrostatic and exchange interactions, and it is strongly R-dependent. If the colliding atoms reach internuclear distances at which the R-dependent term splitting ΔV becomes comparable with the energy defect, $\Delta V \sim \Delta E$, transitions between the terms correlating with different FS components of the $5\,^2D$ state become possible.

The effective cross section for the transition $5\,^2D_{5/2} \rightarrow 5\,^2D_{3/2}$, at the temperature T, can be written as

$$\sigma_{5/2\rightarrow 3/2}(T) = \frac{\sum_i \sum_f g_i \sigma_{if}(T)}{\sum_i g_i}, \tag{8}$$

where summing is done over all the molecular states $|i\rangle$ correlating with the initial $5\,^2D_{5/2} + 5\,^2S_{1/2}$ configuration and all the states $|f\rangle$ correlating with the final $5\,^2D_{3/2} + 5\,^2S_{1/2}$ configuration; $\sigma_{if}(T)$ is the cross section for transitions from state $|i\rangle$ to $|f\rangle$; g_i is the statistical weight of state $|i\rangle$. In deriving Eq. (8), the principal assumptions are (i) the molecular states which correlate with the initial state of separated atoms are populated according to their statistical weights; (ii) the two-level approximation is valid in the calculations of partial cross sections $\sigma_{if}(T)$.

The rotational coupling between the molecular states of different symmetry can be disregarded at large internuclear distances (at $R \geqslant 30$ a.u.) and at thermal collision energies the matrix element of the rotational coupling is small: $V_{\mathrm{Cor}}(R) \sim 10^{-5}$ a.u. Therefore, only the transitions between the terms of the same symmetry induced by radial motion of nuclei are considered.

A characteristic distance R_c is usually introduced for the transition process, based on the relation

$$\Delta V(R_c) = \Delta E, \tag{9}$$

where R_c is determined from the calculated molecular terms. At impact parameters $\rho < R_c$ the inelastic process proceeds in the vicinity of $R \sim R_c$. If $R_c - \rho \gg 1/\gamma$, the transitions in the vicinity of this point can be separated for the collision in an inward and outward direction (as the nuclei approach each other and separate). The parameter γ is determined from the asymptotic behavior of wave function of the valence electron of the excited atom; we use for this parameter the relation $\gamma = \sqrt{2\mathcal{I}_{5D}}$ [21], where $\mathcal{I}_{5D}$ is the first ionization potential of the Rb atom in the $5\,^2D$ state. The transition probability is determined by the Rosen-Zener-Demkov formula [21]:

$$P = \sin^2 \eta / \cosh^2 \delta_R, \tag{10}$$

where $\eta = \int_{-\infty}^{\infty} [\Delta V(R)/2]dt$, $\delta_R = \pi \Delta E / 2\gamma v_R^{\mathrm{rel}}$, and $v_R^{\mathrm{rel}} = v^{\mathrm{rel}} \sqrt{1 - (\rho^2/R_c^2)}$ is the radial component of relative collision velocity v^{rel} of free atoms.

Since, in our case, $\Delta E \ll \frac{1}{2}\,\mu v^{\mathrm{rel}^2}$, where $\frac{1}{2}\,\mu v^{\mathrm{rel}^2}$ is the kinetic energy of the relative motion of atoms, the cross section for the transition $|i\rangle \rightarrow |f\rangle$ can be written [21] in the straight trajectory approximation by integrating Eq. (10) over the impact parameter,

$$\sigma_{if}(v^{\mathrm{rel}}) = \frac{\pi R_c^2}{2} f(\delta). \tag{11}$$

Here $\delta = \pi \Delta E / \gamma v^{\mathrm{rel}}$, $f(\delta) = \int_0^1 [2x\,dx/\cosh^2(\delta/x)]$, and $x = \sqrt{1 - (\rho^2/R_c^2)}$. Equation (11) allows one to determine the cross section for a transition in the two-level approximation to an accuracy of up to $1/\gamma R_c$. Cross section $\sigma_{if}(T)$ is obtained from Eq. (11) by averaging over the Maxwellian velocity distribution:

$$\sigma_{if}(T) = \int_0^{\infty} \sigma_{if}(\mathcal{E}_i) \exp\left(-\frac{\mathcal{E}_i}{T}\right) \frac{1}{T^2}\,\mathcal{E}_i\,d\mathcal{E}_i, \tag{12}$$

where $\mathcal{E}_i$ is the kinetic energy of the relative motion of the nuclei.

Equations (8)–(12) allowed us to estimate the effective cross section for the transition $5\,^2D_{5/2} \rightarrow 5\,^2D_{3/2}$ from Rb_2 molecular terms determined in [8,9]. In the adiabatic Ω basis (Ω is the projection of the total electronic angular momentum onto the internuclear axis), 24 molecular terms arise from the $5\,^2D_{5/2} + 5\,^2S_{1/2}$ and $5\,^2D_{3/2} + 5\,^2S_{1/2}$ atomic states. As mentioned above, we consider transitions between the terms of the same symmetry only, i.e., transitions within the 0_g^+, 0_g^-, 0_u^+, 0_u^-, 1_g, 1_u, 2_g, and 2_u term systems are considered separately. For different terms the value of R_c satisfying Eq. (9) varies in a relatively broad range $32 \leqslant R_c \leqslant 47$ a.u. The value for the parameter δ at mean relative velocity (at $T = 400$ K) is $\delta = 0.4$, which gives $f(0.4) = 0.65$. Calculations according to the above formulas were used for determination of partial cross sections for the terms with $\Omega = 1$ and 2. For the terms with $\Omega = 0$, the above method is not correct because of the avoided crossings with the lower-lying molecular states. For those terms, partial

TABLE I. Rb($5\ ^2D$) FS-changing cross section (in units of $10^{-14}\,\mathrm{cm}^2$).

	$\sigma_{5/2\to3/2}$	
	Experiment	Theory
Present work	5.8 ± 1.9	3.4
Ref. [1]	2.9 ± 0.6	

cross sections were calculated using the results of [9]. In this work, the dynamics of the collisions have been studied in the system of $\Omega=0$ terms.

If the statistical weights of the molecular states are taken into account, then calculation according to Eq. (8) results in the value $3.4\times10^{-14}\,\mathrm{cm}^2$ for the $5\ ^2D$ FS-mixing cross section $\sigma_{5/2\to3/2}$ at the temperature $T=400\,\mathrm{K}$.

IV. DISCUSSION

The theoretical and experimental results for the FS-mixing cross section $\sigma_{5/2\to3/2}$ obtained in this work are compared in Table I with each other as well as with the result $(2.9\pm0.6)\times10^{-14}\,\mathrm{cm}^2$ from [1]. Of the two measured values our result is larger than that of Ref. [1]. Thus, even though our value has been obtained in a cw experiment, this value follows the tendency observed for all results of experiments performed at pulsed excitation by the group of Krause [2–5] for $n\ ^2D$ states with $n=6-9$, if compared with respective results from Ref. [1]. This suggests that the cause of discrepancy between the two series of results, of Ref. [1] and of Refs. [2–5], is different from the possible side effects of pulsed excitation mentioned in Ref. [1].

The theoretical cross section calculated in this work is situated between the two measured values, though closer to the result of [1], lying within its upper limit of uncertainty, and somewhat below the lower uncertainty limit of our experimental value. However, our theoretical value may be underestimated rather than overestimated. The region of intermediate internuclear distances of the Rb_2 quasimolecule can additionally contribute to the process of intramultiplet mixing, and a disregard of this region in the calculation may lead to an underrated FS-mixing cross section. With this in mind, we consider the agreement between the values of our measured and calculated cross sections to be satisfactory.

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