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Collisional transfer within the Sr(5 $^3P^\circ_J$) multiplet due to nearly adiabatic collisions with noble gases

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A time-resolved study of the Sr(5 $^3P^\circ_J$) intramultiplet collisional mixing, following optical excitation of the $^3P^\circ_1$ state, is presented. The degeneracy-averaged rate coefficients of Sr(5 $^3P^\circ_J \rightarrow 5\,^3P^\circ_{J'}$) due to collisions with each of the noble gases in a quasiequilibrium cell are derived from least-squares fittings of the triexponential time behavior of 5 $^3P^\circ_1$ fluorescence and 5 $^3P^\circ_{2,0} \rightarrow 6\,^3S_1$ absorption data. A 5 $^3P^\circ_1$ radiative rate of 22±0.5 μs is also obtained. The dependence of the rates upon buffer pressure demonstrated the presence of both linear and quadratic terms, due to two- and three-body reactions, respectively. The behavior of the binary rate coefficients with varying noble-gas species and temperature is qualitatively consistent with Stückelberg's model for noncrossing levels.

I. INTRODUCTION

Fine-structure-changing transitions within the lowest ^2P doublets of the alkali metals due to collisions with noble gases are a highly studied type of atom-atom interaction.[1] The increase of the alkali-metal fine-structure energy splitting with atomic number provides a large variation of energy defects over which the "two-level" scattering problem can be studied. The lighter alkali-metal atoms Li and Na undergo "sudden" collisions with noble gases, whereas the heavier alkali-metal atoms Rb and Cs represent a nearly adiabatic limit.[1-3] A short discussion of the limits of these two-state models and their conditions of validity appears in Ref. 2. Collisional transfer is said to be sudden or adiabatic depending on the parameter $\xi = (\Delta E/\hbar)/(v/\rho)$ where ΔE is the fine-structure separation, v is the relative velocity of the colliding pair of atoms, and ρ is the effective range of interaction over which level mixing occurs; collisions are sudden when $\xi \ll 1$ and adiabatic when $\xi \gg 1$.

Extensive collisional-transfer data concerning "one-electron" alkali-metal atoms was obtained in the 1960s and 1970s using resonance lamps, primarily by Krause and his collaborators.[1] Similarly complete studies of $^3P^\circ_J$ level mixing in two-valence-electron atoms are only now emerging, as high-power lasers are necessary to adequately excite the partially forbidden $^1S_0$-$^3P^\circ_1$ transition. Experimentally measured state-to-state rate coefficients now exist for the Ca 4 $^3P^\circ_J$ multiplet undergoing collisions with He, and with H_2 and D_2.[4] Total fine-structure-changing cross sections for

$$(\mathrm{Ca}\ 5\,^3P^\circ_1) + M \rightleftarrows (\mathrm{Ca}\ 5\,^3P^\circ_{2,0}) + M$$

have been measured for each of the rare gases, denoted here by M.[5] Theoretical calculations of $^3P^\circ_J$ intramultiplet mixing have been performed for the Mg($3s3p\ ^3P$)-He,[6] and Ca($4s4p\ ^3P$)-He systems.[7] The small energy-level separations of these Mg and Ca $^3P^\circ_J$ multiplets place these collisions in the "sudden" category, and the resulting fine-structure mixing rates are essentially gas kinetic. The group-IIA metals Sr and Ba, which have much larger fine-structure separations, should yield collisional data more characteristic of the adiabatic limit. In essence, the combination of fine-structure and $|m_J|$-dependent interatomic interactions prevents level crossings here, just as in the alkali-metal case. The rate coefficients for level mixing in the heavier group-IIA elements should thus provide a stringent test of the atomic interactions, because such nearly adiabatic transfer is very sensitive to the precise forms of the potential curves and electronic states. This is also true of group-IIB elements Cd and Hg, as confirmed by the small $^3P^\circ_1$ quenching cross sections (due to $J = 1 \rightarrow 0$ transitions) measured for Hg(6 $^3P^\circ_1$) with Xe (Ref. 8) and Cd(5 $^3P^\circ_1$) with Ar and He.[9] However, these studies provided only upper bounds for these cross sections ($< 5 \times 10^{-19}$ cm^2).

Collisional level mixing within the lowest metastable multiplets of group-IIA metals are important for understanding atom-atom collision dynamics, for interpreting lifetime data of some transitions which have astrophysical significance, for understanding metastable-state energy storage, and for studies of chemical reactions involving these excited states.[10]

Collisional mixing of the Sr 5 $^3P^\circ_J$ multiplet has been used to obtain a completely mixed multiplet, whose decay rate yields the radiative lifetime[11,12] and quenching data for the $^3P^\circ_1$ state.[11] These and similar experiments have also obtained diffusion coefficients for Mg, Ca, and Sr ^3P_J-state atoms in buffer gases.[11,13-15] In Refs. 15 and 16 they use an experimental technique similar to that discussed here and they report Ar-induced mixing rates in excellent agreement with our results, as discussed below.

We report here a detailed study of the intramultiplet mixing of the Sr 5 $^3P^\circ_J$ manifold of J states induced by noble-gas collisions. The time-resolved emission of the 5 $^3P^\circ_1$ state prepared by laser excitation, as well as the $^3P^\circ_0$- and $^3P^\circ_2$-state populations measured by absorption, show triexponential behaviors which are analyzed in terms of a coupled three-level rate equation description of populations. This technique of analysis allows accurate determination of state-to-state rate coefficients $k_{JJ'}$(cm^3/s). Unlike Ref. 11, we do *not* observe rapid and complete equilibration of the level mixing at $\sim$700–800 K for Ne, Ar, Kr, and Xe buffer pressures below 100 Torr. However, we do observe rapid J mixing by He ($P > 10$ Torr), consistent with the observation of complete J mixing within $< 10\ \mu$s from Ref. 12.

A large variation of $k_{JJ'}$ with noble-gas species and cell temperatures was observed, which is consistent with an interpretation of these level-mixing rates being due to nearly adiabatic collisions. In addition to the binary-collisional mixing, we have observed intramultiplet mixing proportional to the square of the noble-gas pressure for the heavier noble gases. This unusual behavior has already been reported in a rapid communication.[17]

II. EXPERIMENTAL DETAILS

A. Apparatus

Figure 1 is a schematic diagram of the experimental arrangement. The experiment is performed under quasiequilibrium vapor conditions in a cold-window cell which holds a homogeneous mixture of Sr vapor and buffer gas in the centrally heated region of a stainless-steel cross. An isothermal temperature distribution is maintained within the central 10-cm length of the cross by two resistively heated, tight-fitting brass molds which sheath the $\sim$2-cm i.d. tublar cross, while a cold finger holding the Sr charge (below the plane of Fig. 1) is maintained typically $\sim$20 °C cooler than this central region. The Sr vapor density is approximately determined by the saturated thermodynamic vapor pressure[18] of the cold finger, although the metal vapor continuously condenses to the four transition zones located at the boundaries of the brass mold. In order to minimize energy pooling effects in this study, the excited-state density is generally kept below 10^{11} cm^{-3} by attenuation of the laser beam. The buffer pressures were measured with two capacitance manometers calibrated for 0–10 and 0–1000 Torr with $\pm 0.5\%$ accuracy, and a small mean-free-path to inlet-tube-diameter ratio yields perfect-gas-density relations.

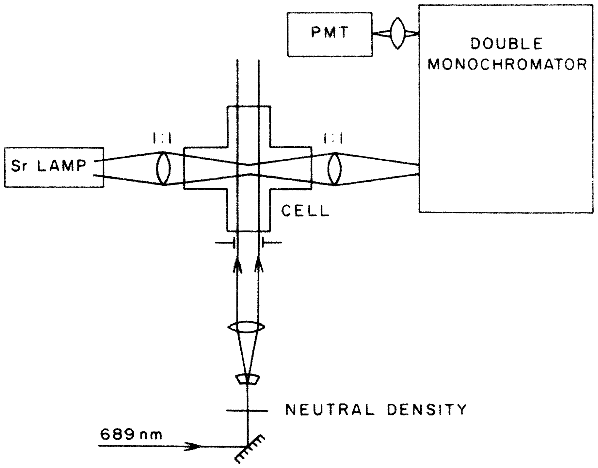

FIG. 1. Experimental arrangement.

The $^3P_1^\circ$ level is excited by a $\sim$0.1 mJ, $\sim$1-cm diameter, $\sim$6-ns pulse from a yttrium aluminum garnet (YAG)-pumped dye laser tuned to the intercombination transition wavelength at 689.2 nm. A Sr hollow cathode discharge lamp is used to perform the absorption studies of the populations of the $^3P_{2,0}^\circ$ levels that result from intramultiplet collisional mixing.

The dye-laser system consists of a single-stage, transversely pumped amplifier and a grazing incidence oscillator which uses a second feedback grating and a prism beam expander to improve its performance.[19] Based on linewidth studies of unsaturated laser-induced fluorescence (LIF) signals, the time-averaged band width of this laser system is $\lesssim 2$ GHz.

We measure a radiative lifetime τ of the $5\,^3P_1^\circ$ state of $22.0 \pm 0.5\ \mu$s, and we have adjusted buffer pressures to produce J mixing in similar times. Thus, the laser pulse width is negligible on this time scale. Complete collisional depolarization occurs on a much faster time scale in our $\gtrsim 1$ Torr buffer, so that isotropic radiation occurs in $< 1\ \mu$s and does not affect the measurements of τ.

The collisional mixing of the initially prepared $^3P_1^\circ$ level with its neighboring $^3P_{2,0}^\circ$ levels is measured directly by studying the time-dependent absorption signals of a dc light source caused by the transient population distribution in the $^3P_J^\circ$ manifold of states. About 10% absorption of the Sr $6\,^3S_1$-$5\,^3P_{0,2}^\circ$ lines from the hollow-cathode lamp can be readily observed through a 12-mm diameter excited-state column density of [Sr($^3P_J^\circ$)]$\sim 10^{10}$ cm^{-3}. Figure 2 shows the levels involved in the excitation and absorptions being observed. A $\frac{3}{4}$-m double monochromator is used to isolate each Sr line whose absorption is studied.

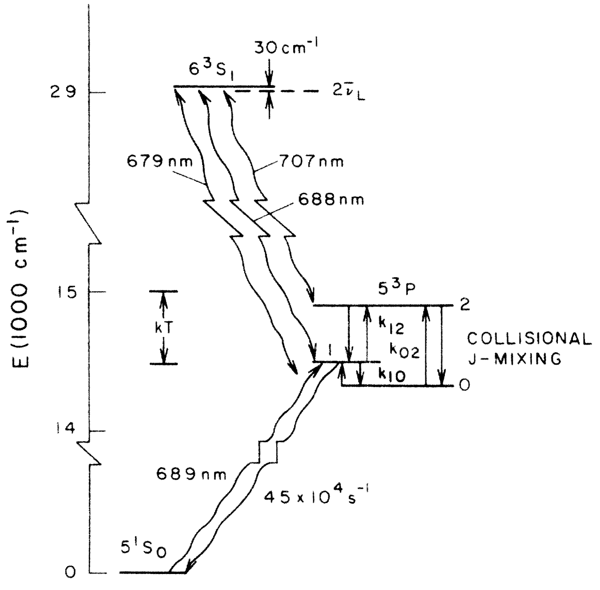

FIG. 2. Sr energy levels relevant to this experiment. The laser at 689 nm excites to the $^3P_1$ state, and the absorption lines at 679 and 707 nm are used to probe the $J=0$ and 2 populations.

The signal acquisition is performed by a 50-MHz-bandwidth, 8-bit (binary digit) resolution transient digitizer coupled to a microcomputer for data storage and analysis. Typically, signals resulting from 2000 laser pulses, at 10 Hz, were averaged to obtain the results presented in Sec. IV.

B. Procedures and corrections

There are several complications to the data analysis. The first results from effects of contaminants that can cause more rapid mixing of the J levels than noble gases (presumably molecular gases which are not quickly gettered by chemical reaction with Sr). In order to obtain meaningful results for the heavier noble gases the partial pressures of such molecular gases must be less than 10^{-6} of the noble-gas pressure. This required extensive bake out of the cell to eliminate water vapor and H_2 trapped in the O-ring window seals and cell walls before consistent results were achieved. That this source of contaminants was adequately eliminated was confirmed by establishing that the J mixing rate was independent of the time after introducing the noble gas. The possibility of molecular contaminants mixed into the reagent-grate noble gases was also checked in the most critical cases of Xe and Kr, by measuring the J mixing rates before and after liquid nitrogen freezing the noble gas into one end of a 1-liter steel bottle and pumping on it to remove more volatile impurities. No difference in the mixing rates was observed after this purification procedure.

A second complication occurs because small but significant initial populations in the $5\,^3P^\circ_{2,0}$ levels are created in the first 50 ns by cascading from the $6\,^3S_1$ level, which is radiatively populated by the laser pulse. This occurs because the pumped $5\,^1S_0$-$5\,^3P^\circ_1$ transition is nearly resonant with the $5\,^3P^\circ_1$-$6\,^3S_1$ transition (the difference is 30 cm^{-1}). Radiative excitation from $5\,^3P^\circ_1$ to $6\,^3S_1$ occurs by absorption in the collision-broadened wing of the transition and also by absorption of the laser amplified spontaneous emission (ASE) at line center. Typically ~10% of the initial $^3P^\circ_1$ population is transferred by J mixing collisions to each of the $^3P^\circ_{2,0}$ levels, while 0.1–1 % of the $^3P^\circ_1$ population is transferred via the $6\,^3S_1$ fluorescence. Thus the initial populations in $^3P^\circ_{2,0}$ produced by $6\,^3S_1$ radiative decay need to be accounted for, since under some conditions they can significantly affect the populations of these levels.

An additional process can transfer excited population between the $5\,^3P_J$ levels. "Energy pooling" by collision of two $5\,^3P^\circ_1$ Sr atoms can produce a ground state Sr and an excited $6s\,^3S_1$ atom, which radiates quickly back to the $5\,^3P^\circ_J$ multiplet. In these studies we have maintained $5\,^3P^\circ$-state densities below 10^{11} cm^{-3} to keep this effect small, and we have measured the J mixing as a function of laser power ($5\,^3P^\circ$ density) to establish the low-power (zero energy pooling) limit.

III. RATE-EQUATION MODEL FOR INTRAMULTIPLET MIXING

As noted above, at the buffer pressures studied complete m_J mixing occurs within the manifold of substates of each $5\,^3P^\circ_J$ level in a very short time compared to the $5\,^3P^\circ_1$ lifetime. Since the depolarization rate is $\gtrsim 10^3$ times the transfer rate, it also occurs very rapidly compared to J mixing. Hence, we will compare the total populations n_J of the three $5\,^3P^\circ_J$ states to the solutions of coupled three-level rate equations, with "initial" conditions representing the three $^3P^\circ_J$ populations immediately (~50 ns) after $5\,^3P^\circ_1$ excitation by the pulse laser and completion of the $6\,^3S_1$ radiative decay to $5\,^3P^\circ_{0,1,2}$. The n_J are then coupled only by collisions, and obey the equations

$$\dot{n}_0 = -(R_{01}+R_{02})n_0 + R_{10}n_1 + R_{20}n_2 \ , \tag{1a}$$

$$\dot{n}_1 = R_{01}n_0 - (R_{12}+R_{10}+\Gamma)n_1 + R_{21}n_2 \ , \tag{1b}$$

$$\dot{n}_2 = R_{02}n_0 + R_{12}n_1 - (R_{21}+R_{20})n_2 \ . \tag{1c}$$

Here $R_{JJ'}$ are the collisional rates for transfer from $5\,^3P^\circ_J$ to $5\,^3P^\circ_{J'}$ and Γ is the natural radiative rate of the $5\,^3P^\circ_1$ state. We reduce the set of six rates $R_{JJ'}$ to three by the detailed balance relation

$$R_{J'J}/R_{JJ'} = (g_J/g_{J'})e^{-[(E_{J'}-E_J)/kT]}$$

A least-squares fitting of the triexponential solutions of these three-level rate equations is performed on the absorption and emission signals yielding values for the parameters Γ, R_{01}, R_{02}, and R_{12}. In principle, one need only perform a single fitting to the absorption or emission signal for any one of the three J states in order to obtain the desired four parameters. However, the parameters are best defined when the least-squares fit is made to all three signals ($J=0,1,2$) at once, and this method was used to analyze the data. Γ was found to have the value 4.55×10^4 s^{-1} ($\tau=22.0\pm0.5$ μs) from these fits, which is about 5% slower than the previously reported values of Refs. 11 and 12. We obtained the same Γ at low buffer pressures (~0.5–5.0 Torr Ar) such that J mixing was reduced as low as possible without diffusion becoming significant. (Radiation trapping effects are not a source of error at the Sr vapor densities used in this work.)

The effects of diffusion and quenching on the J mixing data are expected to be small based on the results of Refs. 12 and 15. That quenching was negligible was experimentally verified by comparing the late-time decay-rate data ($t>250$ μs) with fits based on early-time domain data ($t\lesssim150$ μs). (The early-time data are influenced predominantly by the faster rates of radiative loss and collisional transfer.) Diffusion rates were assessed in the following way: population was produced in the $5\,^3P_2$ state by exciting $5\,^3P_1$ at high laser power, thus enhancing collisional-induced absorption to $6\,^3S_1$, which can then decay to $5\,^3P_2$. At low pressures the population in $5\,^3P_2$ will then be lost via the J mixing rate R_{21} and by diffusion. At 1 Torr Ar, when J mixing is negligible, diffusion was found to occur on a time scale of ~500 μs or $0.05\times\Gamma(5\,^3P_1)$; for the pressures at which J mixing data were taken ($\gtrsim30$ Torr for all gases except He), the effect of diffusion was thus negligible. This procedure also fixed an upper limit for the degree of J mixing at zero pressure, i.e., that caused by collisions of the type Sr*-Sr or Sr*-impurity [R_{21} ($p=0$) $<0.4\times10^3$ s^{-1}]. In the case of He, J mixing is comparable to the radiative rate at much lower pressures (~ 1 Torr), and so a few Torr of Ar were mixed with the He to reduce the diffusion, without producing significant J mixing.

Two examples of fits to experimental data are shown; Fig. 3 for 100 Torr of neon and Fig. 4 for 260 Torr of xenon. The R_{ij} are determined by a least-squares minimization, and the plotted curves represent the best fit under the constraint that the fitting parameters (R_{10}, R_{12},R_{02},Γ) have the same values for all three n_J data sets [the solid line is omitted in Fig. 3(b), as it passes closely through the points]. The absorption for the $J=0$ and 2 levels are related in principle, but minor variations in lamp line profile and pressure shifts cause uncontrolled variations of their ratio. Consequently the n_J signal magnitudes are treated as independent in the fitting procedure. Typical statistical uncertainties from such a fit were typically $\sim 2\%$ for R_{10} and R_{12} and $\sim 100\%$ for R_{02}. This can be understood from the following. The initial decay rate for n_1 is $R_{12}+R_{10}+\Gamma$, so that the high-S/N, $5\,^3P^\circ_1$ fluorescence data accurately determine the sum of these rates. The n_2 data are very sensitive to R_{12} and the n_0 data to R_{10}, as shown in the example fits in Fig. 3(c). This is due to the close connection of the n_2 (or n_0) build-up and emptying rates to R_{12} and R_{21} (or R_{10} and R_{01}). Note, e.g., in Fig. 4 where $R_{10} \sim 2 \times R_{12}$, that the $J=0$ state both acquires and then loses its population more rapidly than the $J=2$ state. In contrast to this, the signal shapes are relatively insensitive to the value of R_{02} due to the initial condition of populating almost entirely $J=1$ and the fact that the various R_{02} were smaller than the other rates by typically a factor of 5. Thus R_{02} has a large fractional uncertainty.

The ratio of "initial" populations of the $^3P^\circ_J$ levels are measured in two ways. In the first method, fluorescence signals out of the $6s\,^3S_1$ state are numerically integrated and adjusted for minor spectrometer plus photomultiplier (PMT) response variations, and normalized to the $5\,^3P^\circ_1$ fluorescence signal to obtain the relative excited-state populations for each $5\,^3P^\circ_J$ level. The "initial" populations of each $5\,^3P^\circ_J$ state were also determined from the least-squares fit to the absorption signal since J mixing occurs on a much longer time scale than the "initial" population build up (50 ns). Both techniques gave values for the initial population ratios n_2/n_1 and n_0/n_1 which matched to within 10%, and as these ratios were typically less than 0.01 this produced 1–5 % uncertainty in the $R_{JJ'}$.

IV. RESULTS

As reported previously[17] the measured transfer rates $R_{JJ'}$ are not proportional to buffer pressure over the range of pressures studied. Clear evidence of a quadratic (three-body collision) contribution to the rates was observed, and this increased with perturber mass. In this work we present and analyze low-pressure data taken to isolate the $R_{JJ'}$ due to two-body collisions. For He, Ne, and Ar the two-body rate is dominant in the pressure range examined, as shown in Fig. 5. The two- and three-body rate coefficients were found by least-squares fitting a linear term plus a quadratic term to these data. The data for Kr and Xe are presented in Ref. 17. For these perturbers the three-body contribution was larger than the

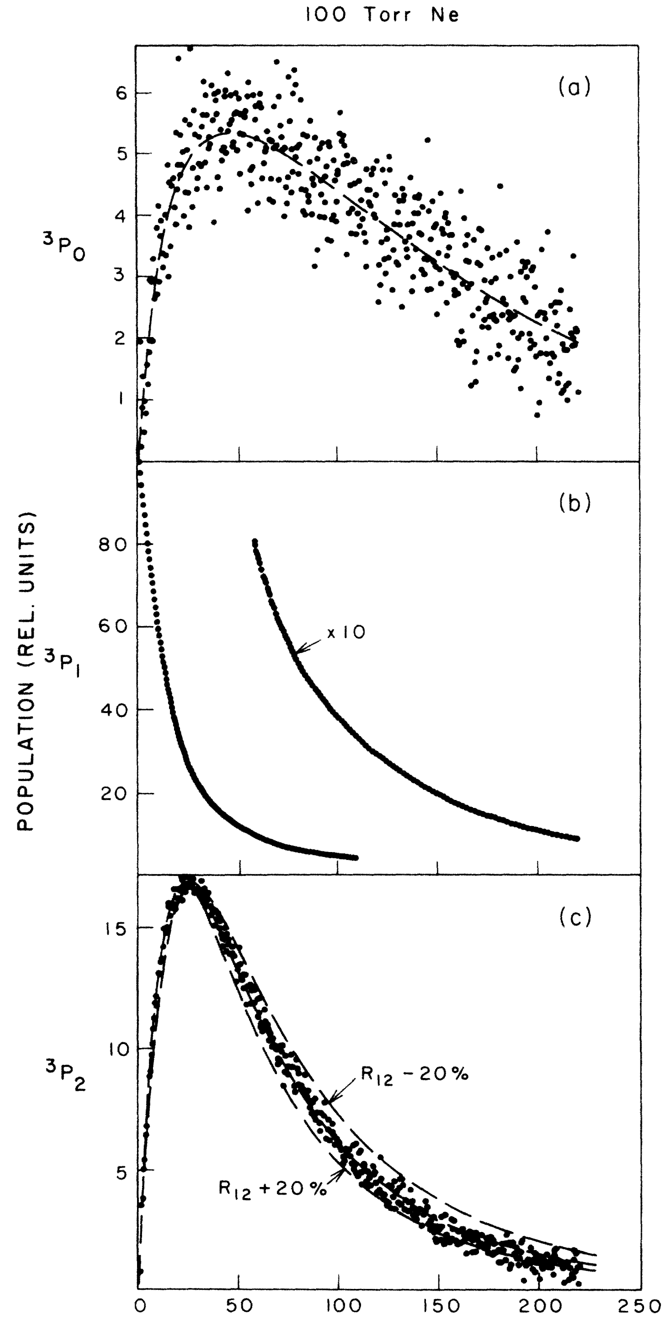

FIG. 3. Time evolution of the $^3P^\circ_{2,1,0}$ populations n_J following the laser pulse for 100-Torr neon: (a) n_2 (from absorption), (b) n_1 (from fluoresence), and (c) n_0 (from absorption). Also shown is the best fit to the data, in which the parameters have the following values: $R_{10}/\Gamma=0.100$, $R_{12}/\Gamma=0.454$, and $R_{02}/\Gamma=0.02$. The relative magnitudes of n_0, n_1, and n_2 are found from the best fit; each data set has been independently normalized to this.

two body at all pressures where useful data could be obtained, and this causes large uncertainties in their measured two-body rates (Table I). As discussed above, the values of R_{02} for all gases were small and ill defined, and their dependence on pressure did not follow any logical pattern. Hence, we only assign upper limits to the values of k_{02}; these are $\lesssim \frac{1}{3}$ of the smaller of k_{12} and k_{10}.

Data were obtained at temperatures ranging from 680 to 810 K in order to assess the temperature dependence of the rate coefficients. The temperature dependence of the measured two-body rate coefficients $k_{JJ'}$ [$=R_{JJ'}$ (linear)/N] are presented in Table I and Fig. 6. Also shown in Table I are the results of Borisov *et al.*[16] for Ar, which are in close agreement with ours.

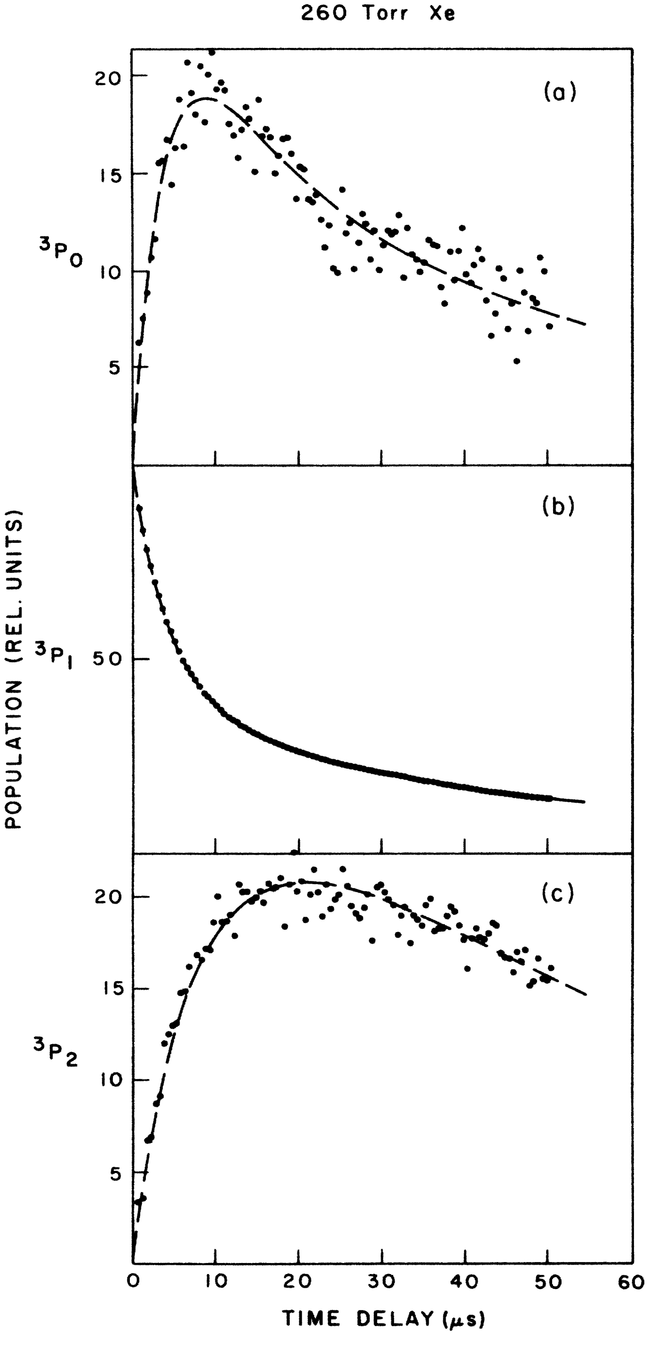

FIG. 4. Same as Fig. 3, but with 260-Torr xenon. ($R_{10}/\Gamma = 1.587$, $R_{12}/\Gamma = 0.928$, and $R_{02}/\Gamma = 0.23$.)

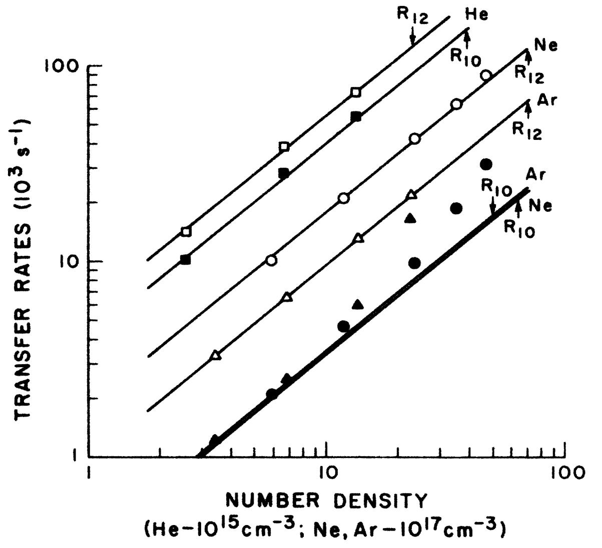

FIG. 5. J mixing rates vs number density for the gases, He (squares), Ne (circles), and Ar (triangles).

The uncertainties in Table I were assessed from the statistical uncertainties in the rate coefficients returned from the least-squares fit of the linear plus quadratic terms to the data in Fig. 5. Several systematic contributions to the uncertainty (discussed in Sec. III) were also included, e.g., from energy pooling, collisional induced absorption to $6\,^3S_1$, quenching, and electronic nonlinearities.

V. DISCUSSION

The J mixing rates measured here for Ne, Ar, Kr, and Xe are all characteristic of highly adiabatic collisions. They are typically 4 orders of magnitude below gas kinetic rates, and are strongly temperature dependent. The

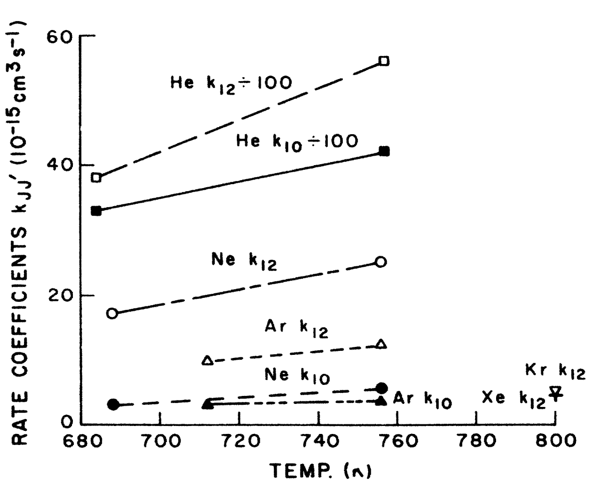

FIG. 6. Measured two-body rate coefficients against temperature (for Kr ∇, Xe +).

TABLE I. Two-body collisional transfer rate coefficients $k_{JJ'}$ for $5\,^3P_J \rightarrow 5\,^3P_{J'}$ in units of 10^{-15} cm^3 s^{-1}. Results of Ref. 16 for $T=700$ K in { }. See text for a discussion of k_{02}.

J,J'	He	Ne	Ar	Kr	Xe
1,0	3300±200 (684 K)	3.0±0.5 (688 K)	3.3±0.5 (712 K) {4±1}	<2.0 (800 K)	<1.0 (800 K)
	4200±100 (757 K)	5.6±0.5 (756 K)	3.8±0.7 (756 K)		
1,2	3800±200 (684 K)	17.1±0.8 (688 K)	9.8±0.5 (712 K) {12±2}	5.0±0.5 (800 K)	4.5±0.7 (800 K)
	5600±100 (757 K)	25.2±1.0 (756 K)	12.3±0.6 (756 K)		

He rates, while being much larger than the others, are still far below the gas-kinetic limit of "sudden" collisional transfer.

This fine-structure mixing is pedagogically equivalent to the alkali $^2P_{3/2} \rightarrow {}^2P_{1/2}$ mixing that was originally explained by Nikitin.[1] In essence, the $\mathbf{L}\cdot\mathbf{S}$ operator dominates at large R, the $|m_L|$-dependent molecular interaction dominates at small R, and the adiabatic states change coupling schemes in the (R_c) region where these energy splittings are comparable. The collisional motion through this "avoided crossing" region then yields a hopping probability as in the Stückelberg or Landau-Zener approximations. As noted, the extent to which collisional transfer is sudden or adiabatic depends on the parameter $\xi=(\Delta E/\hbar)/(v/\rho)$ where ΔE is the fine-structure separation, v is the relative velocity of the colliding pair of atoms, and ρ is the effective range at R_c over which recoupling occurs. The $4\,^3P^\circ_2$ to $4\,^3P^\circ_1$ fine-structure splitting for Ca is $\simeq 106$ cm^{-1}, and from the observation that the Ca J mixing rate coefficients are nearly gas kinetic,[4] one concludes that $\xi < 1$ for this case. The Sr $5\,^3P^\circ_2$-$5\,^3P^\circ_1$ splitting ≈ 400 cm^{-1} and from our data $\xi > 1$ in this case. This appears reasonable, as the molecular ($|m_L|$) interactions are probably similar. The resulting change, by many orders of magnitude, in the fine-structure mixing is similar to that seen for the alkali metals as ΔE_J ranges from 17 cm^{-1} for Na to 550 cm^{-1} for Cs. However, we observe considerably less decrease in the mixing cross sections Q going from Ne to Xe than is seen for Cs fine-structure mixing.

Stückelberg, or Landau and Zener, have shown that in the adiabatic case with the level separation remaining constant at all R, the cross-section scales as $\sigma \sim \exp(-\xi)$.[2,3] Such cross sections as those presented here should, therefore, be strongly dependent on the temperature or relative velocity of the colliding pair, and show marked variation with noble-gas species (since $v \propto (\mathrm{k}T/\mu)^{1/2}$ where μ is the reduced mass of the collision partners, and since ρ depends on the perturber). We do indeed observe an increasing cross section Q with increasing temperature T, but the T dependence is much weaker here than is observed for Rb and Cs fine-structure mixing. No molecular potentials are currently available for Sr(5^3P)–noble-gas interactions, and we have no explanation of these differences at this time.

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