Measurement of ratios of momentum-transport cross sections for potassium−rare-gases by nulling light-induced drift

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(Received 10 November 1997)

We report measured pressure ratios for mixtures of neon with other rare gases that lead to a cancellation of normal light-induced drift of potassium atoms excited on the D_1 line by a narrow-band laser. These data can provide a stringent test for potassium−rare-gas potential curves. To a good approximation, the pressure ratios that cancel normal light-induced drift are directly related to ratios of cross-section differences. Corrections for collisions that cause fine-structure transitions without randomizing the velocity of the potassium atom are discussed. [S1050-2947(98)06706-7]

INTRODUCTION

In a previous work [1] we described the observation of anomalous light-induced drift (LID) of an atomic potassium vapor in a buffer gas consisting of a mixture of two rare gases. This work followed the observation of anomalous LID in a molecular case [2] and a model explanation of the molecular anomalous LID by Chapovsky *et al.* [3]. In the atomic anomalous LID experiment of [1], the two rare gases showed opposite light-induced drift effects and we could mix them in order to cancel the normal light-induced drift effect. In addition to observing the second-order anomalous light-induced drift effect under these conditions, we also noted that the pressure ratios that cancel normal LID can provide an accurate measurement of certain ratios of cross sections, which could in turn provide stringent tests for calculated or measured potential curves for the atom−rare-gas species involved. We reported the pressure ratios used and the resulting values for the cross-section ratios. While providing a better value for the reported cross-section ratios than any other previous experiments or calculations, these measurements were limited by the pressure measurement apparatus that was in place at the time. We report here on follow-up measurements using a more accurate procedure with improved pressure measurements. We also discuss subtle effects of the preservation of atomic momentum during state-changing collisions that could affect the interpretation of the results in terms of cross-section ratios. These effects could also be used as a sensitive tool for studying the degree of preservation of momentum during state-changing collisional transfer between the excited $4P_{1/2}$ and $4P_{3/2}$ fine-structure levels for potassium−rare-gas collision partners.

Potassium is becoming a more studied atom due to the rise in popularity of high-power single-mode Ti-sapphire ring lasers. The existing calculated potassium−rare-gas potential curves are quite old and we expect that the new interest in potassium should provide an impetus for improved calculations. The data reported here would be a severe test of the quality of theoretical potential curves, in the sense that only very good potential curves will be able to correctly predict these experimental results. Alternatively, these data could be used to constrain semiempirical calculations of the potassium−rare-gas potential curves.

EXPERIMENT

The experimental setup is similar to that described in [1] and is reproduced for convenience in Fig. 1. Potassium atoms drift into the main tube at the T intersection and diffuse out toward either side. The large window volumes at the ends of the main tube act as a sink. Interestingly, after many months of operation, the getter action of the potassium cleaned the cell of reacting impurities to the extent that potassium vapor would build up in the window volumes, interfering with the drift velocity measurement. The lifetime of a potassium atom before reacting with an impurity or sticking permanently to the cell wall was on the order of minutes. We placed steel balls at the bottom of the window volumes so that these regions would again act as a sink for potassium vapor. Some of these experiments were also carried out in a different cell of similar design.
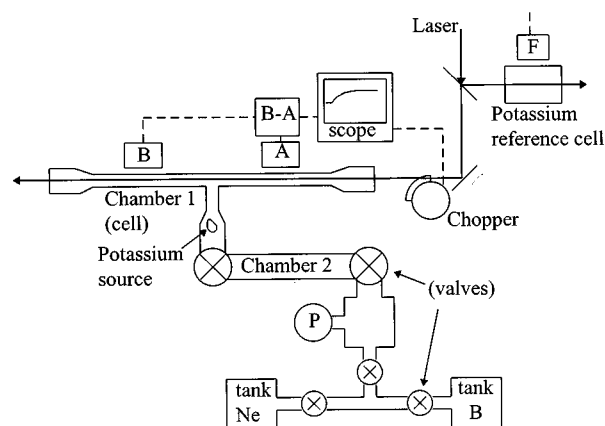

FIG. 1. Experimental arrangement, including a schematic of the vacuum system. Photodiodes are labeled A, B, and F. The box labeled B-A represents a current amplifier that converts the difference current to a voltage for display on the scope. Details of the vacuum system, including the pumps, are not shown.

The laser light propagating in the main tube is tuned 0.25 ± 0.04 GHz to the red of the maximum fluorescence frequency of the D_1 line, measured using a vacuum reference cell shown in Fig. 1. For pure Ne this would cause a light-induced drift pulling effect and for the other rare gases this would cause a pushing effect [1]. If the partial pressures of the Ne–rare-gas mixture are balanced to cancel the light-induced drift, the density of potassium atoms in the main tube is symmetric about the T intersection. The fluorescence entering both detectors shown in Fig. 1 would be the same, resulting in a fluorescence difference signal $A - B = 0$. If there is a slight net pulling effect, the fluorescence difference signal would be positive and a slight pushing effect would produce a negative signal. For small drift velocities, the absolute sign of the signal is subject to errors due to scattered light, amplifier offsets, and imperfections in the symmetry of the cell and detectors, so we chop the laser and look for a time-dependent change over a time scale of about 100 ms. At early times after the beam is admitted, the density profile is still that of zero drift, having formed while the beam was blocked. Then the cloud evolves toward one side or the other according to a positive or negative drift, causing the signal to rise or drop. The displacement of the vapor cloud is larger for larger drift velocities, so that the 100-ms time scale for the cloud to shift is fairly independent of the drift velocity. If the drift velocity is zero, the signal remains flat as a function of time.

The cell is coated with a nonstick coating to prevent the sticking of potassium atoms onto the wall of the main tube. Also, we performed the experiment in a density regime where absorption of the strong laser was small. Experimental details can be found in [1] and references therein.

In Fig. 1 we have included a schematic representation of the vacuum system, as an aid in describing the procedure used for mixing the buffer gases with more accurately known partial pressures. At the relatively low pressures involved (a few Torr) we must exercise care in order to ensure that differences in transport between the different rare gases do not produce errors in obtaining a mixture with well-known partial pressures. The rare gas pressures were measured by a capacitance manometer (MKS Instruments type 122A), which has a specified accuracy of 0.5% reading plus an offset accuracy of 0.01 Torr. This accuracy is a factor of 10 better than the pressure measurements reported in [1]. The ratio of volumes of chambers 1 and 2 in Fig. 1, denoted by R_V, is measured by introducing 100 Torr of one gas (argon) into chamber 1, and letting it expand into the rest of the system (which was evacuated), measuring the final pressure, and repeating the procedure for 100 Torr initially in chambers 1 and 2 together. The ratio of the final pressures is related to the ratio of the volumes in chambers 1 and 2.

The procedure for obtaining a well-characterized mixture in the cell was as follows. First, we introduced a pressure p_1 of gas A into the cell (chamber 1). Then we closed off the valve, evacuated the rest of the system, and introduced a pressure p_2 of gas B into volume 2 and closed off the valve to chamber 2. Then we opened the valve to chamber 1, allowing the gases to mix. The final partial pressures are determined from the original pressures p_1 and p_2 and the ratio of the volumes of chambers 1 and 2. At the low pressures involved, diffusion mixes the gases completely after a few

TABLE I. Pressure ratios that cancel normal LID for total pressure 4 Torr.

Gases AB	P_A / P_B
Ne He	0.32 ± 0.03
Ne Ar	5.25 ± 0.09
Ne Kr	9.5 ± 0.3
Ne Xe	14.4 ± 0.7

minutes. The characteristic mixing time (via diffusion) could be confirmed by watching the drift velocity in the cell change (via the fluorescence difference signal). The observed time scale (~ 20 s) also agreed with an estimate based on a one-dimensional diffusion model, using the approximate cell dimensions. In the previous measurements we tried different pressures until we were close and then diffusively leaked in one of the rare gases until the drift velocity became zero. For these measurements, we kept iterating until we determined a pressure range that gave random results in each direction, thereby determining a more accurate central value, as well as the random component of the uncertainty.

The results for the pressure ratios that balance out the light-induced drift are shown in Table I. Technically, we are measuring the pressure ratio for which the drift velocity is zero at a laser detuning of 0.25 GHz (see the description of the procedure above). This frequency was chosen because it is near the maximum of the drift velocity profile for normal LID, i.e., when a single buffer gas is present. We could have chosen a different measure for assessing when normal LID was ''balanced out,'' for example, by nulling the drift velocity a different detuning or requiring that the drift velocity is minimized for all detunings, but the results would be essentially the same (within the uncertainty) because the drift velocity profile becomes very sensitive to the pressure ratio near the nulling point.

The random component of the uncertainty can be measured statistically from the data and is also in agreement with estimates based on the pressure measurement uncertainties. Systematic uncertainties could arise from the calibration and zero of the capacitance manometer, differences in the transport rates, adsorption of rare-gas atoms into the chamber walls, and the initial measurement of the volume ratio of the chambers. The fluorescence difference signal is extremely sensitive to the pressure ratio, so that the transition from a negative drift to a positive drift is very well defined as a function of the pressure ratio. The determination of the partial pressures is therefore still the dominant source of error. Some of the systematic errors due to transport effects during gas loading can be estimated by changing the order of gases introduced into the cell and observing any discrepancies. These systematic errors also include adsorbing of gas into the tube walls, which is later released when the other rare gas is loaded. The rate of introduction of the gases was also varied in order to check for systematic errors due to differences in transport rates into the chambers and cell. The uncertainties in Table I represent the quadrature sum of these systematic contributions and the observed random component that arises from the pressure measurements. The most

important contributions were from the transport effects and from the random component of the pressure measurements.

DISCUSSION

In [1] we stated that the ratio of partial pressures that cancel out the drift velocity is related to the ratios of cross section differences by

$$\frac{p_A}{p_B} = \frac{\bar{v}_B}{\bar{v}_A} \frac{(\sigma_{eB} - \sigma_{gB})}{(\sigma_{gA} - \sigma_{eA})}, \qquad (1)$$

where $\bar{v}_A = \sqrt{8kT/\pi\mu}$, where kT is the thermal energy and μ is the reduced mass between rare gas A and potassium. This expression is only valid under conditions where no collisional transfer between the excited fine-structure levels is present, or where any collisional transfer between the excited fine-structure levels occurs with complete thermalizing momentum exchange. For the latter case, we define the total momentum transport cross section for each level and for each rare gas by $\sigma_e = \sigma_{ee} + \sigma_{ef}$, where σ_{ee} is the momentum transport cross section for collisions that stay in the excited state and σ_{ef} is for collisions that also change the state to the other fine-structure level [4].

Calculations in which the drift velocity is calculated using a four-level strong collisional model [5] bear out the validity of Eq. (1). However, if we include state changing collisions that do not randomize the velocity, Eq. (1) is not strictly valid. A calculation including a rate for state-changing collisions that do not change velocity, as well as a strong collisional transfer rate described above, shows that the pressure ratio that cancels normal LID is different from Eq. (1). We performed the calculation with parameters modeling the case of potassium in an Ar and Ne mixture, with single-mode excitation tuned near the D_1 line. We chose to use cross sections calculated from the potential curves of Pascale and Vandeplanque [6], using the coupled-channel technique of Hickman *et al.* [4,7] to obtain the various cross sections. We determined a transfer rate for non-velocity-changing collisions, $\Gamma_{n\ i,j}$ (from state i to j), by demanding that the total collisional transfer rate is equal to the sum of velocity-changing and non-velocity-changing rates

$$\Gamma_{\text{tot}\ i,j} = \Gamma_{n\ i,j} + \Gamma_{s\ i,j}, \qquad (2)$$

where $\Gamma_{s\ i,j}$ is the strong collision rate and $\Gamma_{\text{tot}\ i,j}$ is the total collisional transfer rate. Note that in the strong collision model, the strong collisional rate is an effective rate, where several partially randomizing collisions are accounted for by a single randomizing event with a reduced effective frequency. Thus a heavy atom in collision with a light buffer gas would have a much-reduced effective strong collision rate. This model is very good for diffusion and light-induced drift [8]. Although the calculated cross sections based on the Pascale-Vandeplanque potential curves are not adequate for predicting the ratios, we can still use the calculated values to estimate the effects of non-velocity-changing collisions by

TABLE II. Cross section ratios $-\Delta\sigma_A/\Delta\sigma_B = [\sigma_e(\text{Ne}) - \sigma_g(\text{Ne})]/[\sigma_e(B) - \sigma_g(B)]$ calculated from Eq. (1). These ratios are probably accurate to about 4–15 % due to uncertainties from Table I and the effect of non-velocity-changing collisional transfer (see the text).

Gases AB	$-\Delta\sigma_A/\Delta\sigma_B$
NeHe	5.9
NeAr	0.16
NeKr	0.075
NeXe	0.046

comparing the results with the results of Eq. (1). The results show a change in the ratio by 3.5% when the total pressure (Ar+Ne) is held fixed at 4 Torr. For other rare gases, the cross sections have roughly the same magnitudes as for the case of argon. This indicates that we can use Eq. (1) to interpret the pressure ratios from Table I in terms of cross-section differences, with the caveat that the results are probably accurate to within about 4–15 %. (The most accurate case is Ar, for which the most data were collected.) Table II shows the results for these cross-section differences $[\sigma_g(\text{Ne}) - \sigma_e(\text{Ne})]/[\sigma_e(B) - \sigma_g(B)]$, for Ne-$B$ mixtures, where $B =$ He, Ar, Kr, and Xe.

The precise interpretation of the results in terms of ratios of cross-section differences depends on knowledge of the velocity retention during fine-structure-changing collisional transfer. When non-velocity-changing collisional transfer is included, the pressure ratio that cancels normal LID becomes dependent on the total buffer gas pressure. For total pressures ranging to 20 Torr, the effect is on the order of 15%. Given the accuracy of this experiment, we should be able to observe this dependence on total pressure, which in turn would provide information on the non-velocity-changing collisional transfer rate. From this a correction to the results in Table I could be developed, but we defer this analysis for later work.

Currently available potential curves for the potassium–rare-gas interactions do not even get the sign of these fractions right [9]. The measured ratios of cross section differences reported in Table II can be used as a critical test of the quality of more sophisticated calculations of these potential curves as they become available. We challenge any theorist to calculate the potential curves accurately enough to independently predict these results. Alternatively, these data could be used as constraints for semiempirical potential curves. Note that the available potential curves for the potassium–rare-gas interaction for Ne, Ar, Kr, and Xe are from 1974 [6] and for He are somewhat more recent, from 1983 [10]. The availability of stabilized, narrow-band, tunable Ti-sapphire lasers is making potassium an atom of choice among many experimentalists, increasing the need for newer and more sophisticated potential curves.

Given the sensitivity of these measurements to the potential curves, we feel that a relatively unsophisticated transport theory, such as the strong collision model used here and in [1], should suffice for interpreting the pressure ratios in terms of ratios of cross-section differences. For example, a more sophisticated theory may affect the resulting cross section ratios by a few percent, but in any case the results would probably still disagree with calculations based on existing

potential curves by a factor of 2 or more. Thus, even the simple analysis can still contribute significantly to our understanding of the potential curves. A more sophisticated treatment [11,12] may be necessary for future experimental results, such as higher accuracy measurements of the frequency dependence of the anomalous drift velocity or the dependence of these pressure ratios on total buffer gas pressure.

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