

Kr-collision shift of the Rb D_1 transition: The isoclinic point and precision optical spectroscopy

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Measuring the energy dependence of optical-transition collision shifts has proven extremely difficult, in part because of Doppler broadening and the manifold of overlapping hyperfine components that must be disentangled in the spectra. Here, we demonstrate an approach to these measurements based on spectroscopic isoclinic points. To illustrate the approach's efficacy, we investigated the Kr collision shift of the Rb D_1 transition at 795 nm. For the expected Rb-Kr van der Waals interaction, the collision shift should scale like $(T/T_o)^\kappa$, where T_o is a reference temperature and $\kappa_{\text{theo}} = 0.31$. Exemplifying the difficulty of κ determinations, previous alkali-metal–noble-gas experimental measurements of κ have varied widely, sometimes in striking disagreement with theory (i.e., factor of 2 larger). In the present work, we not only demonstrate a measurement precision better than $10^{-10}/^\circ\text{C}$, but with our technique we validate the theoretical scaling constant, finding $\kappa_{\text{expt}} = 0.36 \pm 0.06$.

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I. INTRODUCTION

The shift $\delta\nu$ and broadening γ of optical transitions by perturber atoms and molecules has been an important source of information regarding interatomic potentials for decades [1], and an important special case concerns the broadening and shift of the alkali-metal first resonance lines by the noble gases. In the alkali-metal–noble-gas system, a lone alkali-metal valence electron is perturbed by the closed-shell electronic structure of a noble gas atom. As a consequence of its relative simplicity, this system should provide theory with rich, yet manageable, opportunities for experimental validation, and in some sense this has proven true: Kielkopf has modeled the alkali-metal ground state ($n^2S_{1/2}$) and first excited-state ($n^2P_{1/2,3/2}$) interactions with noble gas atoms via a 6, 8, 10 potential [i.e., $V(R) = -C_6/R^6 - C_8/R^8 + C_{10}/R^{10}$] [2] and has had reasonable success calculating the broadening and shift of alkali-metal first resonance lines [3].

However, one seldom-discussed problem in this otherwise encouraging situation concerns the energy (i.e., temperature) dependence of the collision shift, which remains poorly known and sometimes in striking disagreement with theory [4,5]. As discussed by Peach under the approximation of straight-line trajectories [6], if the interatomic potential can be modeled as $V(R) = \sum C_p/R^p$, then the energy-dependent phase shift per collision, $\eta(E)$, for a specific collisional velocity v and impact parameter b has the form

$$\eta(E) = \sum_p \frac{\sqrt{\pi} \Delta C_p \Gamma[\frac{1}{2}(p-1)]}{v \hbar b^{p-1} \Gamma[\frac{1}{2}p]} = \sum_p \frac{\Delta C_p \beta_p}{v \hbar b^{p-1}}. \quad (1)$$

Here, ΔC_p is the difference in potential coefficients for the alkali-metal's upper, $|2\rangle$, and lower, $|1\rangle$, states: $\Delta C_p \equiv C_{p,2} - C_{p,1}$. The collision shift is then determined by averaging $\eta(E)$ over collisional velocities and impact parameters:

$$\delta\nu = 2\pi N_{BG} \int_0^\infty \left(\int_0^\infty b \sin[\eta] db \right) f(v) v dv, \quad (2)$$

where $f(v)$ corresponds to the Maxwell velocity distribution at vapor temperature T , and N_{BG} is the vapor density of buffer-gas perturbers. If the interaction is dominated by a single term in the potential's power-law expansion, for example, a van der

Waals dipole-dipole term (i.e., $p = 6$), then the integrals can be evaluated in closed form, leading to

$$\delta\nu = \xi_p N_{BG} \left(\frac{\Delta C_p}{|\Delta C_p|} \right) \left(\frac{2|\Delta C_p| \beta_p}{\hbar} \right)^{2/(p-1)} \times T^{(p-3)/2(p-1)} \cos \left[\frac{\pi(p-3)}{2(p-1)} \right], \quad (3a)$$

where

$$\xi_p \equiv \sqrt{\pi} (8k_B/\mu)^{(p-3)/2(p-1)} \Gamma \left[\frac{p-3}{p-1} \right] \Gamma \left[\frac{2p-3}{p-1} \right]. \quad (3b)$$

Here, μ is the reduced mass of the colliding pair, and k_B is Boltzmann's constant. Thus, in general, the collision shift is assumed to have a power-law temperature dependence: $\Delta\nu(T) = \delta\nu_o(T/T_o)^\kappa$, with T_o a reference temperature and κ a parameter sensitively related to the details of the interatomic potential. For the case of alkali-metal–noble-gas interactions, where we expect the interatomic potential to be dominated by a van der Waals $1/R^6$ term, Eq. (3a) predicts that $\kappa \cong (p-3)/(2p-2) = 0.3$.

Unfortunately, with few experimental determinations of κ , it has been difficult to assess the accuracy of these theoretical expectations concerning the dynamics of the collisional perturbation (i.e., its energy dependence). In particular, a recent estimate of κ for the Cs/Ar system ($6^2S_{1/2} \rightarrow 6^2P_{1/2}$ transition) found $\kappa_{\text{expt}} = 0.56 \pm 0.07$ [7], while κ_{theo} for this system is 0.31 [3]. More disturbing than the near factor of 2 discrepancy with theory is the fact that replacement of this κ_{expt} value into the simple expression $(p-3)/(2p-2)$ yields $p = -16$. Specifically, if we assume that a single term dominates $V(R)$, then this κ value suggests that the term scales like R^{16} , which is clearly unphysical. Thus either theory is missing something important in modeling the alkali-metal–noble-gas interaction, or a better approach to κ determinations is needed.

In part, the difficulty in determining κ experimentally arises because the alkali-metal first resonance actually corresponds to a manifold of hyperfine transitions, as illustrated in Fig. 1(a). Consequently, to determine this spectrum's center-of-mass shift, one needs to *accurately account for the overlapping wings* of the individual Voigt line shapes. This is a nontrivial

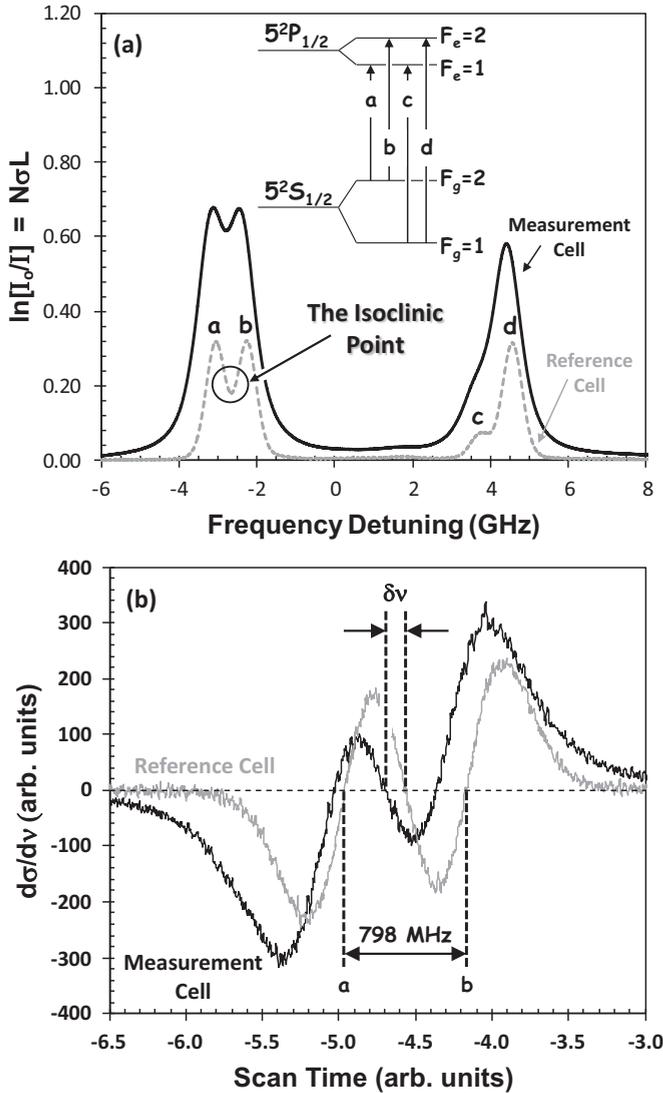


FIG. 1. (a) Scan of the absorption cross section. We measure the transmitted light intensity as a function of the laser frequency and through Beer’s law evaluate $N\sigma L = \ln[I_0/I]$, where N is the Rb number density, σ is the absorption cross section, and L is the cell length. Note that the integral of the absorption profile yields NL , so that by varying temperature in our experiments, the logarithm of the integrated absorption signal should scale like $1/T$; this was verified. (b) Numerical derivative of $N\sigma L$ from (a).

problem, since temperature not only shifts the manifold’s center of mass, it also changes the Doppler broadening of the overlapped resonances, which causes the peaks within the manifold to shift relative to one another. Compounding this problem is the fact that the temperature dependence of the pressure shift is actually quite small. For example, the pressure shift for the D_1 transition of ^{133}Cs , perturbed by 75 Torr of Ar at 313 K, is -485 MHz [7], which leads to $d[\delta\nu]/dT \cong -0.5$ MHz/ $^\circ\text{C}$ (assuming $\kappa = 0.31$). Thus, given the 9193 MHz width of the hyperfine transition manifold in ^{133}Cs , measuring $d[\delta\nu]/dT$ corresponds to finding this manifold’s true center-of-mass shift at the relative level of $5 \times 10^{-5}/^\circ\text{C}$.

Clearly, determining κ is a very difficult task, fraught with the potential for small but significant systematic errors, which

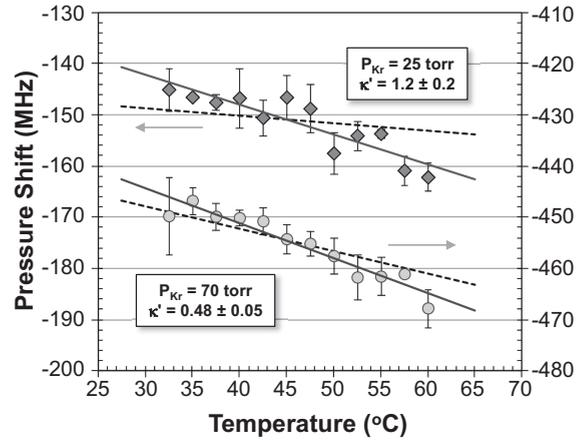


FIG. 2. Pressure shift, as a function of temperature, of the resonant frequency corresponding to the $^{87}\text{Rb } 5^2S_{1/2}(F_g = 1) \rightarrow 5^2P_{1/2}(F_e = 2)$ transition for two different Kr pressures. The dashed lines correspond to theory taking $\kappa = 0.31$. From these data, one would infer κ' values that not only disagree with theory, but that also suggest a pressure sensitivity for κ .

helps explain why so few measurements of κ exist and why there can be greater-than-quoted uncertainty in the measured values. To illustrate the difficulties more concretely, Fig. 2 shows the shift in the peak of the $|F_g = 1\rangle \rightarrow |F_e = 2\rangle D_1$ transition of ^{87}Rb [i.e., transition d in Fig. 1(a)] as a function of temperature for two different noble gas pressures. (We will discuss how these data are generated subsequently.) Of all the resonances in the manifold, this is the least perturbed by pulling from its overlapped neighbor. Nevertheless, not only do these data yield κ values (i.e., κ') in disagreement with theory, but the data suggest that κ' has a pressure dependence, which is in no way consistent with Eqs. (3). Clearly, even though transition d is the nearest thing to an isolated resonance in the manifold, it is not isolated enough to avoid significant systematic errors in κ determinations. To be clear, we are not suggesting that past work has extracted κ values from the data without attempting to control for systematic biases. Rather, Fig. 2 is simply meant to illustrate the sensitivity of the results to those biases, in particular, the temperature-dependent pulling of resonance peaks due to changes in Doppler broadening for overlapped resonances.

In this work, we discuss a technique for determining $d[\delta\nu]/dT$ based on spectroscopic isoclinic points, in particular, the isoclinic point in the D_1 spectra of nuclear-spin $I = 3/2$ alkali metals. Due to the equality of the D_1 matrix elements for the $|F_g = 2\rangle \rightarrow |F_e = 1\rangle$ and $|F_g = 2\rangle \rightarrow |F_e = 2\rangle$ transitions in $I = 3/2$ alkali metals [i.e., transitions a and b in Fig. 1(a)], the midpoint between these transitions (i.e., the isoclinic point) does not shift as a result of changes in line broadening. In particular, barring real energy-dependent effects like pressure broadening, Wells and Camparo [8] have shown theoretically that the isoclinic point has an essentially zero temperature coefficient and (for the $^{87}\text{Rb } D_1$ transition at 795 nm) have measured the isoclinic point’s temperature coefficient as less than 0.002 MHz/ $^\circ\text{C}$. In contrast to the hyperfine manifold’s center of mass, the isoclinic point is an easily observed resonant feature in the absorption spectrum. Further, the isoclinic point will only shift with temperature

to the extent that the collision shift varies with temperature, thereby providing a unique capability for κ measurements [9].

II. EXPERIMENT

A. Overview

Figure 3 shows our experimental arrangement: a vertical-cavity surface-emitting laser (VCSEL) is split into two beams, each of which has a diameter of 0.4 cm and is separately attenuated using a rotating polarizer. One beam with a power of 113 nW passes through the reference cell, which contains only a vapor of ^{87}Rb maintained at 35 ± 0.01 °C. The other beam with a power of 138 nW passes through the measurement cell, whose temperature T is varied between 32.5 °C and 60 °C, and which contains a mixed ^{87}Rb -Kr vapor. The cell diameters are 2.5 cm and their lengths are 4.4 cm, and we employ low laser powers to limit distortions of the spectra due to optical pumping [10]. Our VCSEL laser linewidth is 150 MHz (FWHM).

We employed four different measurement cells in our experiments, each containing different Kr pressures: 0, 24.9, 70.6, and 98.2 Torr. (The 0-Torr cell was included to test for the presence of systematic effects, and for ease of reference, these cells are referred to as our 0, 25, 70, and 100-Torr Kr cells, respectively.) The Kr pressures in the closed cells were measured using the technique of Kazantsev *et al.* [11]. Briefly, the Kr cells were taken to a separate atomic-clock-system setup and employed as the “test clock’s” resonance cell [12]. The frequency shift of the test clock, relative to a GPS-disciplined commercial Rb atomic clock, is dominated by the hyperfine transition’s pressure shift [13] and is therefore a measure of the Kr pressure in the cell. In separate measurements we verified that this technique has an absolute accuracy better than ± 0.2 Torr and a measurement repeatability of $\sim \pm 0.01$ Torr.

B. Measurement procedure and systematic effects

Figure 1(a) shows the absorption cross section in our two cells as a function of laser tuning: the measurement cell contained 25 Torr Kr at 42.5 °C. The absorption cross section, $\sigma(\nu)$, was estimated using Beer’s law: $N\sigma(\nu)L = \ln[I(\nu)/I_o(\nu)]$, where $I(\nu)$ is the transmitted light intensity and $I_o(\nu)$ is the light intensity reaching the photodetector

in the absence of the vapor at laser frequency ν ; this was estimated by tuning the laser both above and below resonance, and fitting the off-resonance transmitted light intensity to a quadratic in ν . Figure 1(b) shows the numerical derivative of these spectra near the isoclinic point. Using the reference cell, we determined the time difference between the zero crossings corresponding to the a and b transitions: δt_{ab} . Knowing the frequency spacing between these transitions $\delta\omega_{ab}$, we could calibrate time change to frequency change. Nominally, this spacing should correspond to the excited-state hyperfine splitting of 812 MHz [14]. However, due to the overlap of the partially resolved absorption line shapes, the observed cross-section peaks are pulled towards one another so that their actual spacing was 798 MHz. This spacing was determined by fitting the measured spectra to two Voigt line shapes. (Notice that the 812 \rightarrow 798 MHz change in the observed excited-state hyperfine splitting only corresponds to a 1.7% difference, implying that any *error* in the calibration due to improper modeling of the overlapped resonances is fractions of a percent.)

We then measured the time interval between the isoclinic points in the two cells’ spectra, $\delta\nu(t)$. Briefly, for the derivative spectrum in each cell, we found the approximate location of the isoclinic point. We then fit the derivative curves about this approximate location to high-order polynomials in order to find the derivative’s zero crossing, varying the temporal range of the fit and the order of the polynomial. (Variability in these numerically extracted zero crossings contributes to our reported error.) From the average difference in the two cells’ zero crossings $\delta\nu(t)$, we could determine the collisional frequency shift of the isoclinic point in the measurement cell: $\delta\nu(\omega) = \delta\nu(t) \times (\delta\omega_{ab}/\delta t_{ab})$. Notice that our calibration procedure determines $d\omega/dt$ over the (limited) isoclinic-point region of the full manifold, so that any (potential) nonlinearities in the laser’s frequency scan over the manifold have an attenuated influence on our frequency shift results. Each scan of the laser frequency was independently “self-calibrated” in this fashion.

One potentially problematic systematic effect in our technique concerns untold asymmetry in the amplitudes of the a and b transitions. Asymmetry could arise from errors estimating $I_o(\nu)$ at different laser tunings, optical pumping [15], or a mixing of hyperfine levels through the alkali-metal–noble-gas spin-orbit interaction [16]. For the overlapped a and b transitions, an amplitude asymmetry implies that the isoclinic point is pulled from its true midpoint frequency. Since this pulling will have a temperature sensitivity, due to the differential effects of Doppler broadening at the isoclinic point arising from the asymmetry, it could potentially give rise to systematic errors in the determination of κ .

To estimate the influence of any asymmetry in our spectra on determinations of κ , we first defined an asymmetry parameter α ,

$$\alpha \equiv \frac{2(A_b - A_a)}{A_b + A_a}, \quad (4)$$

where A_j is the amplitude of the a or b spectral line, and we then examined our spectra for the presence of nonzero α . Focusing on the 25-Torr Kr cell, we measured α as a function

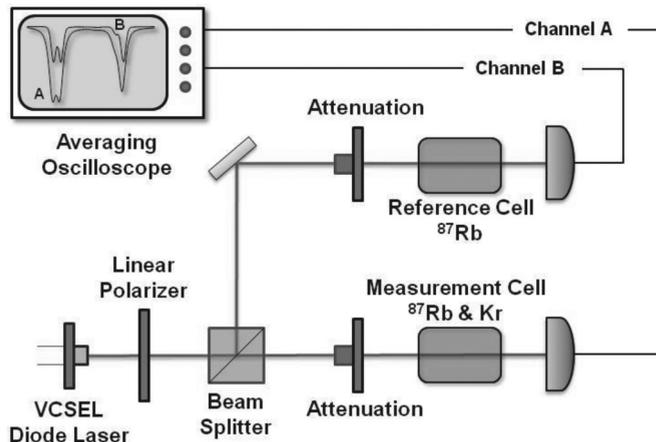


FIG. 3. Experimental arrangement.

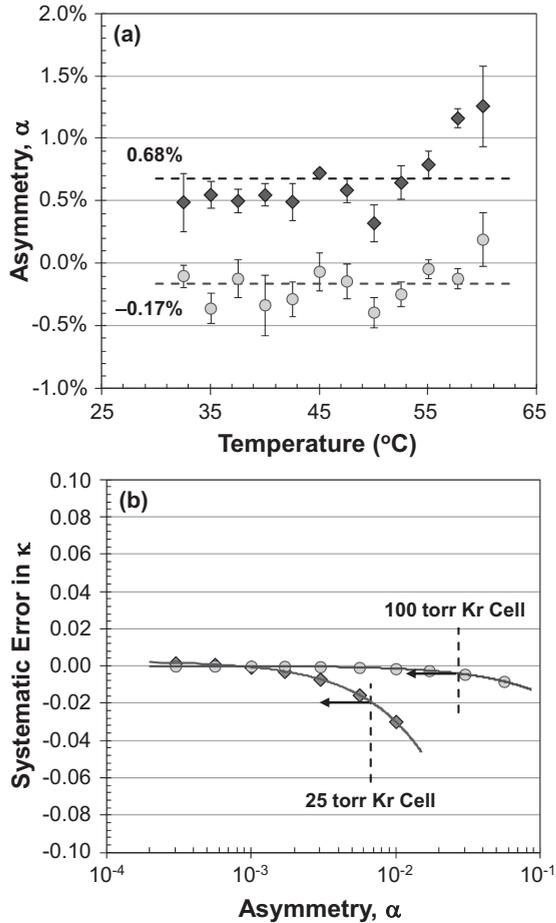


FIG. 4. (a) The asymmetry parameter α as a function of measurement-cell temperature for our 25-Torr Kr cell (diamonds) and our “vacuum” reference cell (circles). (b) Estimate of the asymmetry-induced systematic error in κ as a function of α .

of temperature, and these results are shown in Fig. 4(a), along with the values of α measured in our reference (“vacuum”) cell. (Note that while we plot α as a function of temperature for the reference cell, its temperature was not changing; these reference-cell data were obtained when the measurement cell was at the indicated temperature.) The first thing to note from the data is that the reference cell and measurement cell have differently signed nonzero values of α . This is consistent with an α dominated by errors in the determination of $I_o(\nu)$ at the a and b resonant frequencies. Additionally, for the measurement cell, there appears to be an increase in α with temperature, which is likely a manifestation of an optical pumping effect. Regardless of the asymmetry’s underlying cause, in our 25-Torr Kr cell, averaging over temperature, we have $\alpha \cong 7 \times 10^{-3}$.

Though we can readily measure α in the 25-Torr cell, assessments of α in higher-pressure cells becomes a bit trickier, since the excited-state hyperfine splitting is not resolved. We can note the following, however. If α is dominated by errors in $I_o(\nu)$, then we expect α (on average) to be essentially the same for all measurement cells in our experiment. However, if α arises from the alkali-metal–noble-gas spin-orbit interaction, mixing hyperfine levels, then we should expect α to be roughly

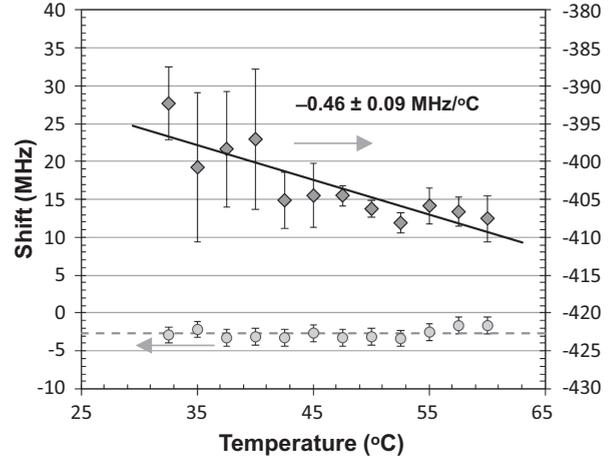


FIG. 5. The shift of the measurement cell’s isoclinic point relative to the reference cell’s isoclinic point as a function of the measurement cell’s temperature: circles \Rightarrow 0-Torr Kr measurement cell, diamonds \Rightarrow 70-Torr Kr measurement cell.

four times larger in our 100-Torr Kr cell. Similarly, if α arises from an optical pumping effect, we expect α to be a function of noble gas pressure. Taking a worse case situation, where increasing buffer-gas pressure improves the efficiency of optical pumping by limiting the rate of diffusion to the measurement cell’s walls [17], and recognizing that optical pumping must already be weak given our low laser intensity, we can assume that in our 100-Torr Kr cell, α will again be 4 times larger than in our 25-Torr cell.

Using Whiting’s first approximation for a Voigt profile [18], we next computed the full D_1 absorption spectrum of ^{87}Rb and determined the shift in the isoclinic point with temperature (in the absence of the pressure shift’s real temperature dependence) for different values of α . This is an estimate of the systematic shift in the isoclinic point with temperature: dv_{sys}/dT . From dv_{sys}/dT , we can determine a κ value, and this is taken as the systematic error in κ due to line-shape asymmetry. This error is plotted in Fig. 4(b), and we see that it is relatively small. Taking the true value of κ as 0.3, Fig. 4(b) suggests that at worst (i.e., in our 25-Torr Kr cell) the systematic error will be less than 7%. Since this is a good deal smaller than the random error in our measurements, as will be reported subsequently, we will ignore systematic errors arising from line-shape asymmetry in what follows.

For each cell, the collision shift as a function of temperature was measured several times, and an example of our data is shown in Fig. 5. The circles in Fig. 5 show the results for the 0-Torr Kr cell. Ideally, there should be no difference in the isoclinic points for this cell and the reference cell, since each has a zero Kr pressure. However, the data show the 0-Torr measurement cell exhibits a $-2.7 \pm 0.3 \text{ MHz}$ shift relative to the 0-Torr reference cell. This shift corresponds to the quantization limit associated with our oscilloscope’s digitization of the time scale: one digitized time unit of the oscilloscope trace corresponded to a frequency change of 2.9 MHz. Thus the 0-Torr cell data of Fig. 5 result from a one time-unit lag between the measurement cell’s time scale and the reference cell’s time scale. Note that this bias is

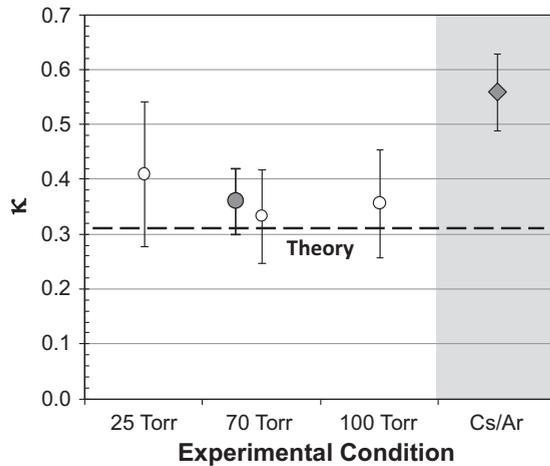


FIG. 6. Various estimates of κ for the D_1 transition: open circles \Rightarrow the individual Kr cell results, filled circle \Rightarrow a weighted mean of our results, diamond \Rightarrow the Cs/Ar D_1 κ value from Ref. [7]. Error bars correspond to 1σ .

temperature independent and therefore has no effect on the κ determinations. Nevertheless, all the data were corrected for this 2.9-MHz bias.

III. RESULTS

Diamonds in Fig. 5 correspond to the collision shift of the 70-Torr Kr cell as a function of temperature after correction for the bias. The temperature variation of the collision shift is readily apparent and leads to $d[\delta\nu]/dT = -0.46 \pm 0.09$ MHz/°C. The isoclinic-point frequency shift versus temperature data of Fig. 5 can also be used to estimate the Rb-Kr D_1 collision shift at our midpoint temperature, $T_o = 318$ K. Specifically, using the data from our 25-Torr Kr cell, which has the most clearly observed isoclinic point as a consequence of pressure broadening, we found $d\nu/dP = -5.01 \pm 0.03$ MHz/Torr, which is a value in very good agreement with the measurements of Kaliteevskii *et al.* obtained at 325 K (i.e., -5.2 ± 0.1 MHz/Torr) [19].

Using data like that shown in Fig. 5, we plotted $\ln[\delta\nu/\delta\nu_o]$ vs $\ln[T/T_o]$ for each of our cells, where $\delta\nu_o$ is the estimated pressure shift at our midpoint temperature. (Note that by taking a ratio of shifts, the time-to-frequency calibration factor falls out of the κ determination.) These plots were linear and yielded κ as the slope. The results are shown in Fig. 6. Open circles correspond to the κ values determined for each of our cells, and the filled circle is a weighted average [20]: $\kappa = 0.36 \pm 0.06$; for completeness we also show the measured κ value for the D_1 transition in the Cs/Ar system (diamond) [7], which theoretically has the same κ value as the Rb-Kr D_1 transition. The dashed line corresponds to κ_{theo} using Kielkopf's 6-8-10

potential [3]. Clearly, our measurements of κ for the Rb-Kr system are in good agreement with theory but contrast sharply with κ_{expt} for the Cs/Ar system. In all fairness, we want to emphasize that the experiments of Kozlova *et al.* [7] were only *peripherally* focused on determinations of κ and were only reported by them for completeness; the main results of their work, the pressure shift of the Cs ground-state hyperfine transition by Ar, remains valid and unquestioned.

IV. SUMMARY

In the present work, we have demonstrated the utility of spectroscopic isoclinic points for precision spectroscopy. Using a very simple and self-calibrating experimental setup, we were able to measure temperature shifts of optical transitions to better than 30 kHz/°C. (This number is based on the 0-Torr Kr data of Fig. 5.) Clearly, this sensitivity could be improved by better attention to the absorption signal-to-noise ratio, and/or direct modulation of the laser frequency for zero-crossing determinations, and/or more data across the temperature range. As a demonstration of the technique's capability, we accurately measured the temperature dependence of the D_1 collision shift in the Rb-Kr system. Contrary to what might be inferred from the experimental literature, we find no evidence of anything seriously lacking in the current theoretical understanding of alkali-metal-noble-gas interactions: our κ_{expt} value agrees quite well with κ_{theo} .

Not only does our isoclinic-point technique provide an improved capability for κ measurements, one can think of turning the problem around. Specifically, if we consider the results from our 70-Torr Kr cell shown in Fig. 5, the measured slope corresponds to an optical fractional-frequency collision shift of $-1.2 \times 10^{-9}/\text{°C}$. However, over a 1 min time interval a diode laser can assess an optical fractional-frequency change at the level of $\sim 5 \times 10^{-12}$ [21]. Thus one could imagine constructing an "isoclinic-point thermometer" for vapor phase systems based on the collision shift: a diode laser would be locked to an alkali-metal isoclinic point, and long-term deterministic frequency variations of the laser would be taken as a measure of temperature-induced changes in the collision shift. Based on the data of Fig. 5, this could potentially provide an atomic and *in situ* capability for monitoring and/or controlling vapor temperatures at the millidegree level every minute. An investigation of the viability of this isoclinic-point application is currently in progress.

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