Two-photon photoassociative spectroscopy of ultracold 88Sr

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We present results from two-photon photoassociative spectroscopy of the least-bound vibrational level of the X1Σ+g state of the 88Sr2 dimer. Measurement of the binding energy allows us to determine the s-wave scattering length, \(a_{88} = -1.4(6)\) \(a_0\). For the intermediate state, we use a bound level on the metastable 1S0−3P1 potential, which provides large Franck-Condon transition factors and narrow one-photon photoassociative lines that are advantageous for observing quantum-optical effects such as Autler-Townes resonance splittings.

I. INTRODUCTION

Precise knowledge of interactions between ultracold atoms has enabled spectacular advances in the production and study of quantum gases. The most accurate tool for determining these interactions is spectroscopy of bound molecular states, such as two-photon photoassociative spectroscopy (PAS) in which two laser fields couple colliding atoms to a weakly bound state of the ground molecular potential via a near-resonant intermediate state (Fig. 1). Two-photon PAS has been used to measure binding energies in Li, Na, K, Rb, Sr, He, and Yb. Each of these measurements provides accurate determination of the atomic s-wave scattering length \(a\) and understanding of the path towards quantum degeneracy and behavior of resulting quantum fluids. Here, we report two-photon PAS of 88Sr and determination of \(a\) for the ground molecular potential (X1Σ+g). Through mass-scaling, we also determine \(a\) for all stable-isotope collisional combinations.

Alkaline-earth atoms such as Sr, and atoms with similar electronic configuration, differ significantly from alkali-metal atoms that are typically used in ultracold experiments. They have a closed-shell ground state structure, numerous isotopes including spinless bosons, and metastable triplet levels that lead to novel laser-cooling techniques and interactions. They present many new opportunities for the study and application of ultracold atoms, such as optical frequency standards, long-coherence-time interferometers, and Bose and Fermi quantum degenerate gases and mixtures. While the collisional properties of these atoms have been the subject of intense study, until now precise scattering length values have only been published for Yb.

For two-photon PAS of Sr, we utilize an intermediate state that is bound in the 0\(\mu\) potential that corresponds to the 1S0 + 3P1 asymptote at long range. The spin-forbidden 1S0−3P1 intercombination transition at \(\lambda = 689\) nm is weakly allowed due to spin-orbit coupling of the 3P1 state with the lowest-lying 1P1 level. PAS involving an intercombination line can change the ground state scattering length with much lower inelastic loss than when using electric dipole-allowed transitions. Long coherence times are helpful for observing quantum-optical effects.
such as Autler-Toews splittings \[29\] of molecular levels \[30, 31, 32\], which is closely related to creation of an atom-molecule dark state \[7, 53\] and state-selective production of ultracold ground state molecules \[51, 34, 55\].

II. EXPERIMENTAL SETUP

A. Laser Cooling and Trapping

To perform two-photon spectroscopy, we start with laser-cooled atoms, and the initial cooling and trapping phases of the experiment are similar to previously published descriptions \[11, 21, 26\]. Atoms are trapped in a magneto-optical trap (MOT) operating on the 461 nm \(1S_0^\text{1} - 1P_1^\text{1}\) transition (Fig. 2) and cooled to about 2 mK. There is a decay channel from the \(1P_1\) state, through the \(1D_2\) state, to the metastable \(3P_2\) level with a branching ratio of \(2 \times 10^{-5}\). To increase our sample number, we repump \(3P_2\) atoms by applying a 3 \(\mu m\) laser resonant with the \(3P_2^\text{3} - 3D_2^\text{1}\) transition that returns these atoms to the ground state. The repumped sample of atoms contains about \(3.5 \times 10^8\) atoms.

After this initial MOT stage, the 461 nm light is extinguished and the atom sample is transferred with more than 50% efficiency to a second MOT operating on the \(1S_0^\text{3} - 3P_1^\text{1}\) intercombination line \[8\]. The sample is cooled to 3 \(\mu K\), producing densities of \(10^{12}\) cm\(^{-3}\). The 689 nm light is provided by a master-slave diode laser system that is frequency-narrowed by servo-locking it to a high-finesse optical cavity with the Pound-Drever Hall method to produce a laser linewidth of \(\sim 50\) kHz. Long-term stability is maintained with a \(1S_0^\text{3} - 3P_1\) saturated-absorption cell.

B. Crossed Optical Dipole Trap

To obtain high density and long sample lifetimes for improved two-photon PAS, atoms are transferred to an optical dipole trap (ODT) generated from a 21 W, 1064 nm, linearly-polarized, single-transverse-mode fiber laser. The trap is in a crossed-beam configuration, derived from the first order deflection of an acousto-optic modulator (AOM). The beam is focused on the atoms with a minimum \(e^{-2}\) intensity-radius of \(w=75 \mu m\). It is then reflected back through the chamber to intersect the first beam at 90 degrees and refocused to have the same waist at the atoms. Both beams lie in a plane that is inclined 10.5 degrees from horizontal. The ODT trapping potential is calculated from measured laser beam parameters and the polarizability of the \(1S_0^\text{1}\) state \[37\]. This allows us to determine the sample density profile from the temperature and number of trapped atoms.

The maximum transfer efficiency observed from intercombination-line MOT to ODT for an optimized overlap time of 80 ms between the two is about 15%. This is limited in large part by inelastic collisions induced by 689 nm light. Atoms are initially loaded with a single-beam ODT power of 5 W, which creates a trap depth of about \(U_{\text{max}}/k_B = 25 \mu K\). After the 689 nm light is extinguished, the power is ramped to a final value between 2.5 and 13 W in 20 ms, yielding equilibrium temperatures of between 3 and 15 \(\mu K\). Up to \(20 \times 10^6\) atoms are loaded to yield peak densities on the order of \(10^{14}\) cm\(^{-3}\).

The number of atoms and sample temperature are determined with time-of-flight absorption imaging using the \(1S_0^\text{1} - 1P_1\) transition. The lifetime of atoms in the ODT due to collisions with background atoms is about 2 s.
C. Photoassociation

After the atoms have equilibrated in the ODT, the photoassociation lasers are applied (Fig. 1). Laser $f_1$ is near resonance with a single-photon, free-bound transition to the red of the $^1S_0-^3P_1$ atomic transition. For some studies, this is the only laser applied. For two-photon PAS, laser $f_2$ is near resonance with a transition from the excited molecular bound state to a ground molecular level.

Using acousto-optic modulators, all photoassociation lasers are derived from the master laser that provides the intercombination-line MOT beams (Fig. 2). Both $f_1$ and $f_2$ lasers are coupled into the same single-mode optical fiber with the same linear polarization. More than 85% of the intensity of both output beams is linearly polarized in the vertical direction, perpendicular to the ODT laser polarization. These beams are focused to a location of the atom sample, which is substantially larger than the atom cloud. The powers are monitored but only states of the 0 ground state lack hyperfine structure. As shown in Fig. 1, ground state photoassociative loss. The observed PAS spectrum is related to the loss spectrum. Depending on the specific measurement, one of the lasers is scanned and the number of atoms remaining after the photoassociative interaction time is recorded to obtain the loss spectrum.

III. THEORETICAL DESCRIPTION OF PHOTOASSOCIATIVE LOSS

Photoassociation is monitored by measuring the loss of ground-state atoms from the ODT. This loss is described with a local equation for the evolution of the atomic density

$$\dot{n} = -2Kn^2 - \Gamma n,$$

where the laser-frequency dependence of the collision event rate constant, $K$, determines the spectrum of the photoassociative loss. The observed PAS spectrum is relatively simple because the bosonic isotopes of strontium lack hyperfine structure. As shown in Fig. 1, ground state $^1S_0$ atoms collide on a single $^1\Sigma_g^+$ potential. Four molecular potentials converge to the $^1S_0-^3P_1$ asymptote [17], but only states of the $^3P_1$ and $^3P_1$ potentials are optically excited from the $^3\Sigma_g^+$ potential [20]. At the low temperatures of atoms in the ODT, only slow collisions occur so only $J = 1$ intermediate levels and $J = 0$ and 2 final states are populated.

Photoassociative loss can be analyzed with the theory of Bohn and Julienne [38], which yields

$$K = \frac{1}{h^2 Q_T} \int |S(E_g, f_1, f_2, \ldots)|^2 e^{-E_g/k_BT} dE_g$$

where the partition function is $Q_T = \left(\frac{2\pi k_BT}{h^2}\right)^{3/2}$ for reduced mass $\mu$. In spite of the low temperature, thermal averaging over the collision energy $E_g$ is necessary because of the narrow linewidth of the transition. $|S|^2$ is the scattering probability for loss and its structure depends upon the loss process that is dominant.

In two-photon spectroscopy of alkali-metal systems, the dominant photoassociative loss process is a collision on the open channel of two ground state atoms $(g)$ with total energy $E_g$ leading to loss-producing decay from the excited state $b_1$ with rate $\gamma_1$. (See Fig. 1.) However, in the experiment reported here, $b_1$ is metastable, and there is a concern that loss from the ground molecular state $b_2$ may also be important. The complete vanishing of the photoassociative loss when the lasers are on two-photon resonance from $g \rightarrow b_2$ (Secs. V and VI), however, implies that decay from $b_2$ is negligible for our conditions and loss from $b_1$ dominates. We can express the scattering probability as

$$|S_{1g}|^2 = \frac{(\delta_1 - \delta_2)^2 \gamma_1 \gamma_s/(2\pi)^2}{\left[\left(\delta_1 - \delta_2 - \frac{\gamma_1}{2} + \frac{\Delta_B^2}{\lambda^2}ight)^2 + \left(\gamma_1 + \frac{\gamma_2}{4\pi}\right)^2 (\delta_1 - \delta_2)^2\right]}$$

$$\gamma_1 = 2\gamma_{\text{atomic}}$$ where $\gamma_{\text{atomic}}$ is the decay rate of the atomic $^3P_1$ level, and $\gamma_s(E_g)$ is the stimulated width of $b_1$ due to laser-coupling to $g$, $\gamma_s(E_g) = \frac{2\pi V^2\langle b_1|E_g\rangle^2}{\hbar}$, where we represent $g$ as the energy-normalized colliding state $|E_g\rangle$, leading to the the Franck-Condon factor for the free-bound transition, $|\langle b_1|E_g\rangle|^2$, and $V = d(\Delta_B^2/2\gamma_{\text{atomic}})$ for free-bound laser intensity $I_1$ and molecular dipole matrix element $d$. Note that our $\Omega_{12}$ is the splitting of the Autler-Townes doublet (Sec. V), which differs from the Bohn-Julienne definition of the molecular Rabi coupling [38].

The thermal energy is much greater than the zero-point energy for trap motion, $T >> h\nu_{\text{trap}}/k_B$, so confinement effects are negligible [24]. We also neglect Doppler shift and photon recoil [10], which is reasonable since $T >> T_R$, where the recoil temperature for $\lambda = 689$ nm photons is $T_R = h^2/(k_B\lambda^2m) = 460$ nK. We assume the decay products leave the trap, which is a good approximation for the intermediate levels we use, although it is not for the least bound $n_a$ excited molecular state [20, 39].

The energy integral for $K$ (Eq. 1) is not analytic and must be evaluated numerically. The situation is further complicated by the ODT, which is not at the magic wavelength for one-photon photoassociation (914 nm) [20]. The AC Stark shift of the weakly bound ground molecular level ($b_2$) is approximately equal to the shift of the incoming channel of 2-free atoms $(g)$ [32, 40]. In other words, the polarizability of the ground molecule is about twice that of a single atom. But the ground and excited molecular levels do not experience the same shift. For
spectroscopy, we can thus treat the ODT Stark shifts as a position-dependent shift of the intermediate state and define the laser detunings
\[
\delta_1 = f_1 - (E_{b_1} - E_g)/\hbar - \chi_{ODT}(\vec{r}) \\
\delta_2 = f_2 - (E_{b_2} - E_g)/\hbar - \chi_{ODT}(\vec{r}),
\]
where \(I_{ODT}(\vec{r})\) is the intensity profile of the optical dipole trap and \(\chi\) can be related to the differences in polarizabilities for \(^1S_0\) and \(^3P_1\) atoms for 1.06 \(\mu\)m laser light.

This implies that \(|S|^2\) and thus \(K\) are functions of position, which must be addressed when Eq. 1 is integrated over the trap volume to calculate the time evolution of the number of trapped atoms
\[
N(t) = \frac{N_0 e^{-\Gamma t}}{1 + \frac{2N_0 K_{\text{eff}} V}{N_0^2} (1 - e^{-\Gamma t})},
\]
where \(N_0\) is the number at the beginning of the PAS interaction time. The one-body loss rate, \(\Gamma\), is due to background collisions and off-resonant scattering from the PAS lasers. The effective volumes are defined by
\[
V_q = \int_V d^3r e^{-\frac{V_{ODT}(r)}{\hbar^2}},
\]
where \(U(r)\) is the trap potential, and
\[
K_{\text{eff}} = \frac{1}{V_2} \int_V d^3r e^{-\frac{V_{ODT}(r)}{\hbar^2}} \times \frac{1}{h Q_T} \int_0^{U_{\text{max}} - U(r)} dE_g |S_{1g}|^2 e^{-E_g/k_B T}.
\]
The kinetic energy integral is truncated by the local trap depth, \(U_{\text{max}} - U(r)\). The spatial integrals in Eqs. 7 and 8 extend over the trap volume \(V\) in which \(U(r) < U_{\text{max}}\). Atom temperatures vary by no more than 25% during the interaction time, so assuming a constant sample temperature is reasonable.

The spectrum is sensitive to many atomic and molecular parameters, and multiple types of spectra can be used to determine them. The ultimate goal is an accurate determination of \(E_{b_0}\) because the molecular binding energy determines the \(s\)-wave scattering length and the underlying potential with high accuracy.

IV. ONE-PHOTON PHOTOASSOCIATION

A. One-Photon PAS Spectrum

One photon PAS allows us to determine \(\chi\), the relative light shift of states on the ground and excited molecular potentials (Eq. 5), and the stimulated width \(\gamma_s(E_g)\) (Eq. 10).

When \(I_2 = 0\) and \(\Omega_{12} = 0\) in Eq. 9, we recover the one-photon PAS scattering probability for loss through decay of \(b_1\)
\[
|S_{1g}|^2 = \frac{\gamma_1 \gamma_s/(2\pi)^2}{\delta_1^2 + \frac{1}{(2\pi)^2} \left( \frac{\gamma_1 + \gamma_2}{2} \right)^2},
\]
\[
\text{FIG. 4: Left: Atom number versus free-bound laser detuning from the one-photon } ^1S_0-^3P_1\text{ atomic transition (color online). Spectra shown here are for 6 W and 13 W ODT single-beam powers with sample temperatures of 6 \(\mu\)K and 13 \(\mu\)K, respectively. Right: Collision-event rate constant } K_{\text{eff}} \text{ derived from the atom loss. The ODT at 1064 nm causes an AC Stark shift of the excited molecular state compared to the ground state, which shifts and broadens the line. The solid lines are fits using Eqs. 9 and 10. A peak shift of 480 kHz is measured for a single-beam power of 13 W. The dashed line marks the position of our measured unperturbed resonance frequency at -222.25(15) MHz, which is in reasonable agreement with a previous measurement of -222.161(35) MHz [24].
\]

The state \(b_1\) is equal to the \(J = 1\) rotational state of the third-lowest bound vibrational level of the \(0_u^+\) potential, with energy \(E_{b_1} = E_{3P_1} - h \times 222.161(35)\) MHz [24]. The Condon point for this excitation, where \(V_{0_u}(R_c) - V_{1S_0}(R_c) = E_{b_1}\), occurs at \(R_c = 75 a_0\), which is very near the node in the ground state wave function [24, 41].

Fitting data of atom number after a given interaction time, \(N(t)\), to Eq. 6 yields the collision-event rate constant \(K_{\text{eff}}\). Figure 4 shows typical spectra for this transition.

B. Determining the Stimulated Width

The area under the one-photon PAS line (Fig. 1) can be related to molecular and experimental parameters through
\[
A = \int dE_g K_{\text{eff}} = \frac{1}{h Q_T} \int dE_g e^{-E_g/k_B T} \frac{\gamma_s(E_g) \gamma_1}{\gamma_s(E_g) + \gamma_1}.
\]
Here, we have neglected truncation of the energy integral, which is a small correction. The Wigner threshold law implies \(\gamma_s(E_g) \propto I_1 \sqrt{E_g}\). So for low laser intensity \((\gamma_s(E_g) \ll \gamma_1)\), the expression \(A \approx \frac{k_B T}{h Q_T} \langle \gamma_s(E_g) \rangle\) is
I for find parameter (Eq. 5) can be varied to fit the data (Fig. 4). We one-photon PAS spectra, and the relative light shift parameter allows us to model the bound laser intensity. The area under one-photon PAS spectra versus free-bound laser intensity, showing the polarizability of $\chi^2$ with $[37]$, which yields a peak shift of 480 kHz for our frequency fixed such that $\delta_2 \approx 0$ while scanning $f_r$. The splitting of the spectra is given by the Rabi frequency $\Omega_{12}/2\pi$ and varies as $\sqrt{f_r}$ (Shown by the lines; spectra offset is proportional to $\sqrt{f_r}$), where the bound-bound laser intensity $I_2$ is indicated in the legend. The asymmetry in the line shapes arises from the bound-bound laser frequency being slightly off resonance from the bound-bound transition. The free-bound intensity $I_1$ is constant for all four spectra at 0.05 W/cm$^2$. The sample temperature is 8 µK.

C. Modelling the Spectra and Determining the Relative AC Stark Shift

Numerical integration of Eq. 8 to find $K_{eff}$ using Eq. 9 for the scattering probability allows us to model the one-photon PAS spectra, and the relative light shift parameter (Eq. 5) can be varied to fit the data (Fig. 1). We find $\gamma_f = 160 \pm 30$ kHz/100 kHz/cm$^2$, in good agreement with $[37]$, which yields a peak shift of 480 kHz for our deepest trap. The line shifts to the blue with more ODT laser intensity, showing the polarizability of $^3P_1$ atoms is less than the polarizability of $^1S_0$ atoms.

At higher temperatures, the lineshapes in Fig. 4 possess red tails, which result from the convolution of the Lorentzian spectrum with the Maxwell-Boltzmann distribution of collision energies $[16, 20]$.

V. TWO-PHOTON AUTLER TOWNES SPECTRUM

If PAS spectra are recorded in the same fashion as in Sec. [IV] except the bound-bound laser is added near resonance ($\delta_2 \approx 0$) with a large intensity $I_2$, the loss spectrum is modified due to the coupling between $b_1$ and $b_2$. This forms a $\Lambda$ system and the line is split into an Autler-Townes doublet, with splitting given approximately by $\Omega_{12}/2\pi$, where

$$\Omega_{12} = \alpha |\langle b_1 | b_2 \rangle| \gamma_1 \sqrt{I / 4I_{sat, atom}}. \quad (11)$$

The overlap integral is related to the Franck-Condon factor, $F$, through $F = |\langle b_1 | b_2 \rangle|^2$. The saturation intensity for the atomic $^1S_0 - ^3P_1$ transition is $I_{sat, atom} \equiv \pi h c |\bar{\alpha}|/(3\lambda^3) = 3 \mu W/cm^2$. The rotational line strength factor, $\alpha$, accounts for the change in dipole moment from atom to molecule due to symmetry of wave function and projection on a rotating molecular axis $[17, 19]$.

Figure 6 shows several Autler-Townes spectra for various intensities $I_2$ for a ground molecular state $|b_2\rangle$ equal to the $J = 0$ rotational state of the least-bound ground vibrational level. This is the $v = 62$ level counting from the bottom of the well. We find that $\Omega_{12}/2\pi = 1$ MHz for an intensity of 0.35 W/cm$^2$, which yields $F = 0.28 \pm 0.06$ for $\alpha = \sqrt{2/(3)} \lambda^3 [17]$. The energy of the ground molecular state, $E_{b_2}$, can be found from fits of data in Fig. 6 but it is more accurately found by varying $\delta_2$ with $\delta_1 \approx 0$, as we will discuss in Sec. [VI].
The asymmetry in the line strengths in each doublet in Fig. 6 arises from the coupling-laser frequency being slightly off resonance from the Stark-shifted bound-bound transition ($\delta_2 \neq 0$). But $\delta_2$ is small and the scaling with intensity shows that the Autler-Townes splitting varies as $\sqrt{I_2}$ as expected.

These spectra show the potential of the system for quantum optics and ultracold molecule formation. The condition of no PAS loss when both lasers are on resonance has also been called a dark resonance [33], or an atom-molecule dark state [33] because the system has been proposed as a vehicle for creating large numbers of the ground state potential, as described in Fig. 7 (color online). The free-bound laser frequency is fixed close to the one-photon PAS resonance and the intensity is $I_1 = 0.05 \text{ W/cm}^2$. The bound-bound laser frequency is scanned, and its intensity is indicated in the legend. On two-photon resonance, PAS loss is suppressed due to quantum interference. The solid lines are fits using Eqs. [8] and [3] which yield $E_{b2}/\hbar = -136.7(2) \text{ MHz}$. The atom temperature is $8 \mu \text{K}$.

VI. TWO-PHOTON SUPPRESSION OF PHOTOASSOCIATION

For determining the binding energy of molecular levels of the ground state potential, we hold the frequency of the free-bound laser fixed close to the one-photon resonance, $\delta_1 \simeq 0$, and scan $\delta_2$. When $\delta_2 - \delta_1 = 0$, the system is in two-photon resonance from state $g$ to $b_2$, and one-photon photoassociative loss is suppressed due to quantum interference. At this point, $f_1 - f_2 = (E_{b2} - E_g)/\hbar$, so the spectrum allows accurate determination of $E_{b2}$. An average over $E_g$ is necessary in order to properly account for thermal shifts of the resonance.

Figure 7 shows a series of spectra taken at various bound-bound intensities for $b_2$ equal to the $J = 0, v = 62$ state. Detuning of the free-bound laser frequency ($f_1$) from the free-bound resonance, which depends on the ODT light shift ($\chi$) and collision energy ($E_g$), causes slight asymmetry in the lines and broadening, but this shape is reproduced with our model for $K_{eff}$ (Eq. 8) using parameters independently determined in previous sections. Since the initial and final states experience roughly equal light shifts due to the trapping laser, the ODT AC Stark shifts do not shift the resonance. No significant shift of the binding energy with laser power was observed, and we place an upper limit of 100 kHz for our highest intensity, $I_2 = 0.55 \text{ W/cm}^2$. We have also measured the binding energy of the $J = 2, v = 62$ state (Table I), and a typical spectrum is shown in Fig. 8.

VII. DETERMINATION OF THE SCATTERING LENGTH

Binding energy measurements can be used to accurately determine the $s$-wave scattering length for the $X^1\Sigma_g^+$ ground electronic molecular potential and to ob-

\begin{table}[h]
\begin{tabular}{|c|c|c|c|c|}
\hline

Isotope & $v$ & $J$ & Exp. & Theory & Diff. \\
\hline

88 & 62 & 0 & -136.7(2) & -136.7 & 0.0 \\
88 & 62 & 2 & -66.6(2) & -66.5 & -0.1 \\
\hline
\end{tabular}
\caption{Observed ground molecular levels and experimental and theoretical level energies in frequency units ($E_{b2}/\hbar$).}
\end{table}
tain information on the van der Waals coefficients, $C_n$. For our analysis, the inner part (internuclear spacing $R < 20.8 \, a_0$) of the potential is described with a recently published energy curve [43] derived from the Fourier-transform spectrum of Sr$_2$ and additional information on the zero-energy ground-state scattering wave function from PAS [21]. A multipolar van der Waals expansion in $C_n/R^n$ is used to represent the potential at longer range ($R > 22.7 \, a_0$), and the gap between the two regions is bridged with a spline interpolation to insure a smooth connection. The wave functions are calculated using a full quantum calculation [21].

As was the case in the analysis of [43], we do not have enough information to independently determine all the van der Waals coefficients, and improve on the precise relativistic many-body calculation of [44], which gave $C_6 = 3103(7)$ a.u., $C_8 = 3.792(8) \times 10^5$ a.u., and $C_{10} = 4.215 \times 10^7$ a.u. The last bound level ($v = 62, J = 0$) is very extended, with its outer turning point at roughly $R \sim 100 \, a_0$. At this point, the leading contribution to the dispersion energy, $-C_6/R^6 - C_8/R^8 - C_{10}/R^{10}$, arises from the $C_8$ term. In fact, using the values of $C_n$ above, the $C_8$ contribution is only 1.22% of that of $C_6$, while the $C_{10}$ contribution accounts for only 0.02%. At a shorter distance $R \sim 20 \, a_0$, similar to the maximum separation of states measured in [44], these contributions are roughly 30.5% and 8.5%, respectively. In [44], the precise theoretical values of [44] for $C_6$ and $C_8$ were used to fit the value of $C_{10}$, since the energy levels ($v = 0 - 50$) were more deeply bound and corresponded to shorter range than our levels ($v = 62, J = 0$ and 2). We note that less precise values of $C_6$ and $C_8$ from [43] were also considered in [44].

If we use $C_6$ as a fit parameter to match the binding energy of the $J = 0, v = 62$ state, assuming $C_8$ and $C_{10}$ from [44], the best fit value is $C_6 = 3151(1)$ a.u. where the quoted uncertainty only reflects uncertainty in the measured binding energy (see Table I). The resulting $^{88}$Sr $1S^2 \Sigma^+ g$ s-wave scattering length is $a = -2.0(2) \, a_0$, where the uncertainty also only reflects uncertainty in the measured binding energy. If we use the value of $C_{10} = 6.60 \times 10^7$ a.u. from [44] instead, we find $C_6 = 3116.0(5)$ a.u., in reasonable agreement with [44]. The resulting scattering length is $a = -1.2(2) \, a_0$. If instead, we take $C_6$ and $C_8$ from [44], and fit $C_{10}$, as was done in [44], we find $C_{10} = 7.488 \times 10^7$ a.u. and $a = -0.9(2) \, a_0$ (again, with the uncertainties reflecting the uncertainty in the measured binding energy).

It is difficult to assess the uncertainties in $a$ related to these coefficients. The most conservative assessment encompasses the full range of values quoted here; $a = -1.4(6)$ and $C_6 = 3130(20)$ a.u. The uncertainties in $C_6$ and $C_8$ quoted in [44] are quite small, however, and no uncertainty is quoted for $C_{10}$. So that might give more credence to the results for $C_6$ and $C_8$ from [44] and the resulting fit $C_{10} = 7.488 \times 10^7$ a.u., which corresponds to the higher ends of the ranges of values for $a$ in Table I. Mass-scaling can be used to determine the scattering lengths for all stable-isotope collisional combinations from this information about the potential (Table III).

We note that the rotational energy takes the form $H_{rot} = B(R) J (J + 1)$ where $B(R) = h^2/(2\mu R^2)$ is the rotational constant for separation $R$ and reduced mass $\mu$. The rotational constant for $v = 62$ calculated using this potential yields a binding energy of $-66.5$ MHz for the $J = 2, v = 62$ state (Table I), well within the measurement uncertainty. We also found that this quantity does not significantly constrain the $C_n$ coefficients.

In the ultracold regime, the energy dependence of scattering lengths can often be neglected. However, this is not the case when there is a low-energy scattering resonance or when the zero-energy scattering length is very small. Figure 9 demonstrates that $^{88}$Sr-$^{88}$Sr and $^{86}$Sr-$^{86}$Sr collision cross sections vary significantly with collision energy, even at energies below 1 $\mu$K. This may explain the discrepancy between this work and previous studies of Sr collisional properties [40].

| Isotopes | 2-phot. PAS [this study] | Fourier-transform [43] | 1-phot. PAS [21] | 1-phot. PAS [41] | Thermalization |
|---------|--------------------------|------------------------|------------------|------------------|----------------|
| 88-88   | -1.4(6)                  | 0(5)                   | $10^{+3}_{-1}$   | $-40^{+10}_{-40}$ | 21$^{+3}_{-4}$ |
| 87-87   | 96.2(1)                  | 97(2)                  |                  |                  |                |
| 86-86   | 823(24)                  | 1050(380)              | $1000^{+400}_{-400}$ | $-430^{+80}_{-90}$ |
| 84-84   | 122.7(3)                 | 124(3)                 |                  |                  |                |
| 88-87   | 55.0(2)                  | 56(2)                  |                  |                  |                |
| 88-86   | 97.4(1)                  | 99(2)                  |                  |                  |                |
| 88-84   | 1790(130)                | $> 1170$ or $<-1900$  |                  |                  |                |
| 87-86   | 162.5(5)                 | 165.5(5.5)             |                  |                  |                |
| 87-84   | -56(1)                   | -55(10)                |                  |                  |                |
| 86-84   | 31.9(3)                  | 33(3)                  |                  |                  |                |

TABLE II: Published values of scattering lengths given in units of $a_0 = 0.053$ nm.
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