Systematic study of Optical Feshbach Resonances in an ideal gas

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Using a narrow intercombination line in alkaline earth atoms to mitigate large inelastic losses, we explore the Optical Feshbach Resonance (OFR) effect in an ultracold gas of bosonic 88Sr. A systematic measurement of three resonances allows precise determinations of the OFR strength and scaling law, in agreement with coupled-channels theory. Resonant enhancement of the complex scattering length leads to thermalization mediated by elastic and inelastic collisions in an otherwise ideal gas. OFR could be used to control atomic interactions with high spatial and temporal resolution.

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The ability to control the strength of atomic interactions has led to explosive progress in the field of quantum gases for studies of few- and many-body quantum systems. This capability is brought about by magnetic field-induced Feshbach scattering resonances (MFR) [1], where both the magnitude and sign of low-energy atomic interactions can be varied by coupling free particles to a molecular state. MFR in ultracold alkali atoms have been used to realize novel few-body quantum states and study strongly correlated many-body systems and phase transitions [11, 12]. However, magnetic tuning has limited current experiments to relatively slow time scales and low spatial resolution. Higher resolution could be achieved by controlling MFR optically [3].

Scattering resonances can also arise under the influence of laser light tuned near a photoassociation (PA) resonance [2] where free atom pairs are coupled to an excited molecular state [5, 6]. This Optical Feshbach Resonance (OFR) is expected to enable new and powerful control with high spatial and temporal resolution. OFR has been studied in thermal [7] and degenerate [8, 9] gases of Rb, but it was not found useful due to large photoassociative losses. Much narrower optical intercombination lines are available in alkaline earth atoms and are predicted to overcome this loss problem [10]. Independently, ultracold alkaline earth atoms have recently emerged to play leading roles for quantum metrology [11, 13] where precision measurement and many-body quantum systems are combined to study new quantum phenomena [14, 15]. Degenerate gases of alkaline earth atoms have recently become available [16]. Due to the lack of magnetic structure in the ground state of these atoms, the OFR effect could become an important tool for controlling their interactions. OFR work on Yb [17, 18] has been limited to studying the induced change in scattering phase shifts and PA rates. Dominant PA losses are evident in all of the OFR experiments listed above. Light-induced elastic collisions for thermalization were not observed.

In this Letter, we study the OFR effect across multiple resonances in a metastable molecular potential of 88Sr. The aim of this work is to test the practical applicability of OFR for engineering atomic interactions in the presence of loss, similar to the successful application of a decaying MFR [19]. For 88Sr, OFR is predicted to allow changes in the scattering length by more than a factor of 100 with low losses by using large detunings (O(10^5) linewidths) from the least-bound vibrational level [20]. We tested this proposal and find experimentally that the existing isolated resonance model [4] only describes the experiment in the small detuning regime. Large detunings from a molecular resonance require a full coupled-channels description of the molecular response. Supported by this new theory framework, we present a systematic experimental study of the OFR-enhanced complex scattering lengths and demonstrate OFR-induced thermalization in an ultracold gas.

Bosonic 88Sr has an s-wave background scattering length a_{bg} = -1.4(6)a_0 [21], where a_0 is the Bohr radius. The small |a_{bg}| makes the sample effectively non-interacting and provides an ideal testing environment for OFR. Figure 1 shows the ground \(^{1}S_0 \rightarrow^{1}S_0\) and lowest excited state \(^{1}S_0 \rightarrow^{3}P_1\) molecular potentials of 88Sr, which are coupled by a PA laser near the atomic transition at \(\lambda_s = 689\) nm. The vibrational levels investigated are labelled by their quantum number n, counted as negative integers from the free particle threshold. For a given PA laser detuning from threshold, the Franck-Condon principle localizes the atom-light interaction in the vicinity of the Condon point [6].

When detuning the PA laser across a vibrational resonance, the s-wave scattering length shows a dispersive behavior, just as for a MFR. However, the finite lifetime of the excited molecular state leads to loss intrinsic to OFR. This process can be described [22] akin to decaying MFR [1, 11] with a complex s-wave scattering length \(a(k) = a(k) - i\ell_b(k)\) that depends on the relative momentum \(hk\) and a PA line strength factor \(\ell_{opt} = \frac{\lambda_0}{\ell_b} \left| \langle n(E) \right| \right| k \rangle\), called the optical length [10, 23]. Here, \(c\) is the speed of light, and \(\ell_{opt}\) scales linearly with PA intensity I and free-bound Franck-Condon factor \(\left| \langle n(E) \right| \right| k \rangle\) per unit collision energy \(E = h^2k^2/(2\mu)\) at reduced mass \(\mu = m_{Sr}/2\). In the isolated resonance approximation [3] the inelastic
collision rate is

\[ K_{\text{in}}(k) = \frac{4\pi \hbar}{\mu} \frac{\ell_{\text{opt}} \gamma_{\text{m}}}{\gamma} \frac{(\Delta + E/k_B)^2}{2} \left[ 1 + 2k \frac{\ell_{\text{opt}} \gamma_{\text{m}}}{\gamma} \right]^2 / 4, \]

(1)

where \( \Delta \) is the laser detuning from molecular resonance [22]. We have accounted for extra molecular losses with \( \gamma > \gamma_{\text{m}} = 2\gamma_{\text{a}} \), where \( \gamma_{\text{m}} \) is the linewidth of the molecular transition and \( \gamma_{\text{a}} = 2\pi \times 7.5 \) kHz is the atomic linewidth. Neglecting \( a_{\text{bg}} \) for \(^{88}\)Sr gives \( K_{\text{in}}(k) \approx 2k \frac{\ell_{\text{opt}} \gamma_{\text{m}}}{\gamma} K_{\text{in}}(k) \). The elastic-to-inelastic collision ratio \( K_{\text{el}}/K_{\text{in}} \) becomes less favorable for smaller \( k \).

We load \( \sim 5 \times 10^4 \) atoms from a magneto-optical trap operating on the \(^1\Sigma_0^+\)^3P_1 intercombination transition into a crossed optical dipole trap formed by tilted horizontal (H) and vertical (V) beams (1064 nm), with trap depths \( \sim 15 \) \( \mu \)K and \( \sim 7 \) \( \mu \)K, respectively. The trapped sample shows a clear kinetic energy inhomogeneity between the H and V axes (2-2.5 \( \mu \)K vs. 3-4 \( \mu \)K), due to the negligible \( a_{\text{bg}} \), consistent with a thermal distribution energy-filtered by the trap potential. Typical in-trap cloud diameters are 45-55 \( \mu \)m. The PA beam intersects the trap with a waist of 41 \( \mu \)m [22].

A representative survey of PA resonances in the \(^1\Sigma_0^+\)^3P_1 \( 0_u \) potential is shown in Fig. 1b. The PA laser with intensity \( I \), adjusted to achieve similar \( \ell_{\text{opt}} \) for all spectra shown, interacts with the sample for \( \tau_{\text{PA}} \). Photon-atom scattering at rate \( \Gamma_{\text{sc}} \) and subsequent radiation trapping set the maximum usable \( I \) for a given detuning from the atomic line [22]. In addition to the vibrational levels indicated in Fig. 1a, the \( n=1 \) vibrational state exists at -0.4 MHz detuning from the threshold, which leads to a PA resonance with a very large line strength \( \ell_{\text{opt}}/I [20] \). The isolated resonance theory indicates that operating with a large \( I \) at \( \mathcal{O}(10^5 \gamma_{\text{m}}) \) detuning from the \( n=1 \) state should allow modifications to \( a(k) \) of \( \mathcal{O}(100 a_0) \) [10]. This prediction relied on extrapolating the large line strength of the \( n=1 \) state across multiple intermediate PA resonances. However, with \( I \) up to 1 kW/cm\(^2\) and detunings up to -1.5 GHz, we did not observe any effects due to elastic collisions.

The discrepancy between theory and experiment stimulated a coupled-channels treatment of an atomic collision in a radiation field that properly switches between the short range molecular states and two field-dressed separated atoms [22,24,25]. In the coupled-channels theory, the two coupled excited potentials \( (0_u, 1_u) \) have the form of Ref. [20], with an added imaginary term \(-i\hbar \gamma_{\text{m}}/2\). The ground state potential uses the dispersion coefficients of Ref. [20], has a scattering length of \(-1.4 a_0\), and reproduces the bound state data of Ref. [21] to better than 0.4%. Coupled-channels calculations do not assume isolated resonances, and all \( 0_u \) and \( 1_u \) molecular eigenstates emerge from the calculation as interfering, decaying scattering resonances [1].

Figures 2a and d show that the coupled-channels model reproduces the isolated resonance expressions [1,6] for \( a(k) \) and the rate constants as long as \( \Delta \) is small compared to the spacing between molecular levels. However, the coupled-channels \( K_{\text{el}} \) returns to its background value \( K_{\text{bg}} \) in between resonances regardless of their relative strengths. The dotted line indicates \( K_{\text{el}}(a_{\text{bg}}) \) at \( E/k_B = 4 \) \( \mu \)K in Fig. 2a (Fig. 2d). These calculations show that each molecular line behaves as an isolated resonance near its line center. For detunings comparable to the molecular level spacing, the isolated resonance expressions cannot be used.

At intermediate detunings, \( |\Delta| \gg \gamma(1 + 2k\ell_{\text{opt}} \Delta/\gamma) \), \( a(k) \) can be written in the standard form for MFR [22],

\[
\lim_{k \to 0} a(k) = a_{\text{bg}} \left( 1 - \frac{w}{\Delta} + \frac{i}{2} \frac{w \gamma}{\Delta^2} \right),
\]

(2)

where \( w \equiv -\ell_{\text{opt}} \gamma_{\text{m}}/a_{\text{bg}} \). To obtain a meaningful change in scattering length, \( \ell_{\text{opt}} \gamma_{\text{m}}/\Delta \) needs to be sufficiently large, and the imaginary part \( b = \frac{i}{2} \ell_{\text{opt}} \gamma_{\text{m}}/\Delta^2 \) needs to be sufficiently small. Since \( K_{\text{in}} \approx (2 \times 10^{-12} \text{cm}^3/\text{s}) (b/a_0) \), for a density of \( \rho = 10^{12} \text{cm}^{-3} \) and \( b = 0.1 a_0 \), \( K_{\text{in}} \rho = \Gamma \) for \( I = 53 \) \( \text{W/cm}^2 \) assumed for Figs. 2a,d. Thus, the calculations predict that changes in the scattering length of order 10 \( a_0 \gg |a_{\text{bg}}| \) should be possible with \( \mathcal{O}(100 \gamma_{\text{m}}) \) detunings on timescales of 200 ms.
To investigate the utility of OFR, we systematically characterized three different resonances and determined their universal scaling. Because $K_{\text{el}}/K_{\text{in}} \propto k\ell_{\text{opt}}$, inelastic collisions dominate the dynamics of the sample for small $\ell_{\text{opt}} \ll (2k)^{-1} = \hbar \sqrt{\pi/8mk_BT}$, where the angled brackets indicate a $k$-average at temperature $T$, and $k_B$ is the Boltzmann constant. In this regime, the result of scanning the PA laser across resonance is a loss feature that shows no dependence on elastic collision processes.

A typical PA loss feature for small $\ell_{\text{opt}}$ is shown in Fig. 3, where the final atom number after application of PA light is shown with respect to PA detuning from $^1S_0-^3P_1$. The per-axis kinetic energies [22] for this scan correspond to a horizontal (vertical) temperature $T_H(T_V) = 2 \mu K$ ($3 \mu K$), resulting in the typical thermal tail towards the red side of the resonance [27]. The solid line is a result of solving [28]

$$\dot{\rho}(x,t) = -2K_{\text{in}}(x)\rho^2(x,t)/2 - \rho(x,t)/\tau_{\text{fg}},$$

with single particle density $\rho$, thermally-averaged inelastic collision rate $K_{\text{in}}(x) = (K_{\text{in}}(k,\Delta, x))$ [22], and vacuum-limited trap lifetime $\tau_{\text{fg}} \approx 1.3$ s. Equation 3 is solved at each $x$ and we integrate the density to obtain the number of atoms at the end of the PA pulse. We then use Eq. 3 to fit to the experimental data [22] and extract $\ell_{\text{opt}}$ and the position of the line center. Figure 3 shows the time dependence of the same photoassociative loss process. Two-body loss is evident under resonant PA light.

From the experimental data we extract two independent quantities: $\ell_{\text{opt}}\gamma_m$ and an increased molecular loss rate $\gamma \simeq 2.7\gamma_m$. We have ruled out magnetic field or PA laser noise as a source of broadening. Instead, we conclude that this extra broadening is related to a faster molecular decay rate, which is consistent with our earlier measurements [20] and Rb results [8].

The measurements were performed for a range of $\ell_{\text{opt}}$ by adjusting $I_{av}$. Multiple molecular resonances were measured and results for $n = 2$ are shown in Fig. 3. The optical length data is fit with a linear function and the results are summarized in the table at the bottom. The fit coefficient $\ell_{\text{opt}}/I_{av}$ is given by the free-bound Franck-Condon factor and decreases drastically with decreasing $n$. Figure 3 exemplifies similar measurements done to determine the line shift $\Delta\nu$ with $I_{av}$. Linear shift coefficients $\Delta\nu/I_{av}$ and zero intensity line positions $\nu_n$ with respect to the atomic transition are also shown in the table. The sign and magnitude of $\Delta\nu/I_{av}$ are consistent with the predictions in Ref. [23].

At larger optical lengths ($\ell_{\text{opt}}\gamma_m/\gamma \approx 100a_0$), elastic
collisions start to influence the dynamics of the system. We show the atom loss with respect to PA laser detuning for \( n = 2 \) in Fig. 4. Both in-trap size and kinetic energy are measured by absorption imaging \[22\]. Far detuned from the resonance, the gas is almost ideal, as shown by the persistent kinetic energy inhomogeneity along H and V in Fig. 4. On resonance (vertical dashed line), inelastic collisions dominate and cause heating. For red detuning from the molecular resonance, the temperatures approach each other by cross-thermalization \[24\].

The measured cloud widths \( w_H \) and \( w_V \) confirm that the potential energy follows the kinetic energy (Fig. 3) since particles oscillate in the trap many times between collisions. Similar measurements were performed for \( n = 3 \) and \( n = 4 \), and we find that the same dispersive behavior in temperatures and widths appears around \( 2\langle k^{2}\rangle_{\text{opt}}\gamma_{m}/\gamma \sim 30\% \) at \( \tau_{PA} = 200\, \text{ms} \). The data can be understood by a simple picture of competition between \( K_{el} \) and \( K_{in} \), which average differently with \( k \) in a thermal sample [see Eq. (4)], and thus peak at different values of \( \Delta \). Elastic collisions cause cross-dimensional thermalization and tend to equalize \( T_H \) and \( T_V \). Since inelastic collisions predominantly remove cold atoms from the densest part of the cloud, the resulting loss increases the average system energy via anti-evaporation.

This behavior is confirmed by a Monte-Carlo simulation, where \( 55 \times 10^3 \) particles are simulated and each particle undergoes elastic and inelastic collisions with an initial phase-space distribution matched to the experimental conditions \[22\]. The solid lines overlaid on the experimental data in Fig. 4 are the simulation results. An average ratio of elastic to inelastic collisions per particle from the simulation is shown in Fig. 4. The dispersive shapes are also predicted by the coupled-channels model (see Fig. 2) and their shape is sensitive to \( \gamma \). Combined with the low \( \ell_{\text{opt}} \) data in Fig. 3, the entire simulation reproduces the experimental data only when \( \gamma = 3 \pi \times 40(5) \, \text{kHz} \) without other free parameters.

We conclude that the isolated resonance approximation universally describes OFR in the vicinity of each resonance. The coupled-channels calculation includes all interference effects between resonances, and differs from the isolated resonance approximation at large detuning between resonances. Our experiment contradicts previous predictions based on extrapolations of an isolated resonance to large detunings \[10, 20\]. We have validated the linear line strength scaling and linear resonance shift with \( I \) and have observed a clear modification of both \( a(k) \) and \( b(k) \). For the values of \( \ell_{\text{opt}}\gamma_{m}/\gamma \) achieved here, inelastic losses still contribute significantly and \( \langle K_{el}/K_{in}\rangle \) becomes even less favorable with decreasing \( T \). However, the OFR effect can modify interactions in a degenerate gas of alkaline earth atoms and the desired change of \( a(k) \) is achieved at the smallest \( \Delta/\gamma \) constrained by both molecular and atomic loss processes over a given experimental timescale \[22\].

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SUPPLEMENTARY MATERIAL

Scattering matrix, cross sections, and collision rates

The collision of two identical bosons in the low energy near-threshold limit can be described in a scattering matrix treatment by a single s-wave scattering matrix element, $S(k) = e^{i2η(k)}$, where $\hbar k$ is the relative momentum of the collision pair and $η(k)$ is the scattering phase shift due to interactions. When there is loss of scattering flux from the entrance channel, as is the case of a decaying optical Feshbach resonance, it is convenient to define a complex energy-dependent scattering length $α(k)$, related to the complex phase $η(k)$ and S-matrix element by [1, 2]

$$α(k) = a(k) - ib(k) = -\frac{\tan η(k)}{k} = \frac{1}{i\hbar k} \Gamma + S(k). \tag{4}$$

This reduces to the usual definition of complex scattering length as $k \to 0$. The respective elastic and inelastic s-wave collision cross sections are [2, 3]

$$σ_{el}(k) = \frac{2π}{k^2} |1 - S(k)|^2 = \frac{8π α(k)^2 f^2(k)}{k}, \tag{5}$$

$$σ_{in}(k) = \frac{2π}{k^2} (1 - |S(k)|^2) = \frac{8π}{k} b(k) f(k),$$

where $f(k) = [1 + k^2|α(k)|^2 + 2kb(k)]^{-1} \to 1$ as $k \to 0$ and $μ$ is the reduced mass of the pair. The second relations in Eq. 5 follow from the definition in Eq. 4. The corresponding collision rate coefficients are

$$K_{el}(k) = \frac{\hbar k}{μ} σ_{el}(k), \tag{6}$$

$$K_{in}(k) = \frac{\hbar k}{μ} σ_{in}(k).$$

When the entrance channel is coupled by a single frequency laser to a single excited molecular bound state of the pair, the isolated resonance approximation for the field-dressed s-wave scattering matrix element can be written as [4, 5],

$$S(k) \simeq e^{2iη_0(k)} \left(1 - i\frac{Γ_s(k)}{E/ℏ + Δ + i(γ + Γ_s(k))/2} \right). \tag{7}$$

Here $E = ℏ^2 k^2 / 2μ$ is the collision energy with $μ = m_{Sr}/2$. The laser detuning $Δ$ from the PA resonance includes the light shift [3] to simplify the presentation (see Eq. 20 for a full expression).

The stimulated width $Γ_s(k) \simeq 2kℓ_{opt}(k)γ_m$ is the induced coupling between the free particle state $|E⟩$ and the excited molecular state $|n⟩$. Here $γ_m = 2γ_a$ is the linewidth of the molecular transition and $γ_a = 2π × 7.5$ kHz is the atomic linewidth. We have allowed for extra molecular losses by letting $γ > γ_m$. The optical length

$$ℓ_{opt} = \frac{λ_0^2}{16πc} \frac{|⟨n|E⟩|^2}{I}, \tag{8}$$

where $c$ is the speed of light, is a PA resonance-dependent line strength parameter [3, 7]. Under the Wigner threshold law, the free-bound Franck-Condon factor per unit collision energy $|⟨n|E⟩|^2 ∝ k$, and we checked numerically that it is a very good approximation to take $ℓ_{opt}$ to be independent of collision energy for temperatures $< 10 μK$. The optical length is also independent of the atomic oscillator strength, and it scales linearly with PA intensity $I$.

Combining Eq. 7 with Eqs. 4-6 gives the near-threshold isolated resonance approximation to the OFR inelastic loss rate coefficient:

$$K_{in}(k) = \frac{4πℏ}{μ} \frac{γ_m}{Γ_m} \frac{Γ_{opt}}{γ} \frac{ℓ_{opt}γ_m}{(Δ + E/ℏ)^2/γ^2 + [1 + 2kℓ_{opt}γ_m]2/4}. \tag{9}$$

The background s-wave collision length $a_{bg}$ is defined in the absence of a resonance as

$$a_{bg} = -\lim_{k \to 0} \frac{\tan η_{bg}(k)}{k}. \tag{10}$$

If we neglect $a_{bg}$ for $^{88}$Sr, we find a simple expression for the OFR-induced elastic collision rate coefficient for an ideal ultracold gas

$$K_{el}(k) \simeq 2kℓ_{opt}γ_m^{κ_2} K_{in}(k). \tag{11}$$

Power broadening is included via $Γ_s(k)$ in the denominator of Eq. 7, which corresponds to $2kℓ_{opt}γ_m$ in the denominator of Eq. 9. From the expressions for elastic and inelastic collision rates, we see that the relevant strength parameter in the presence of extra molecular
loss is the rescaled optical length $\ell_{\text{opt}} \gamma_m$. Elastic collisions and power broadening only become important for the dynamics when

$$2k \frac{\ell_{\text{opt}} \gamma_m}{\gamma} \sim 1.$$  \hspace{1cm} (12)

**Coupled Channels Calculations**

The simplest way to do a calculation of the elastic and inelastic collision properties of an optical Feshbach resonance is to set up a coupled channels model for the collision of field-dressed states in a laser field of fixed frequency $\omega$. We use the simplest three-channel model that is sufficiently complete to represent nonperturbatively the optically induced $S(k)$ for an interfering spectrum of excited state resonances. One channel represents the ground state with potential $V_g(R)$ and two channels represent the two excited states 0 and 1 that correlate with the separated atom limit $^1S_0$ and $^3P_1$ respectively complex potentials $V_{0u}$ and $V_{1u}$. These potentials are shifted to include the asymptotic detuning of the laser frequency from atomic resonance. It is necessary to include both $0_u$ and $1_u$ excited states to correctly calculate the complex scattering length near an isolated resonance of either state. This is because the molecular $J = 1$ excited states ($J =$ total angular momentum quantum number) must become a mixture of $s$ and $d$-waves at long range to properly go to separated atoms with $^3P_1$ quantized in a space frame instead of a molecular body frame $^3$: otherwise, spurious $1/R^3$ resonant dipole terms would get mixed into the ground state potential and give invalid threshold scattering lengths.

The coupled channel potential matrix $V(R)$ is found using the asymptotic representation of the two excited $J = 1$ molecular channels in terms of a pair of excited state $s$- and $d$-waves for the $e$ parity block in Table 1 of Reference $^3$:

$$V = \begin{pmatrix} V_g & V_{\text{opt}} \\ V_{\text{opt}} & \frac{1}{2}(V_{0u} + 2V_{1u}) \frac{\sqrt{2}}{3}(V_{1u} - V_{0u}) \frac{1}{3}(2V_{0u} + V_{1u} + 6B) \end{pmatrix},$$  \hspace{1cm} (13)

where $6B = 6\hbar^2/(2\mu R^2)$ is the $d$-wave centrifugal energy. Here the row labels, in order from top to bottom, represent the $|j\ell JM\rangle = |0000\rangle$ ground state and $|1010\rangle$ and $|1210\rangle$ excited state channels of Ref. $^3$, where $j$ is the atomic angular momentum for the collision partner to the $^1S_0$ atom, $\ell$ is partial wave, and $JM$ represent the total angular momentum and its projection for the pair of interacting atoms. Since the excited bound states are relatively short range, we use the nonretarded optical coupling $V_{\text{opt}} = (2\pi I/c)^{1/2} d_m$, where $d_m = \sqrt{2} d_A$ and the atomic transition dipole $d_A = 0.086816$ atomic units corresponds to an atomic lifetime of 21.46 $\mu$s. A complete theory would use the retarded optical coupling. The zero of energy for ground state collisions is taken to be the lowest eigenvalue of this matrix to account for the energy shift of the field dressed molecule. The numerical $S$-matrix is calculated using standard coupled channels methods to calculate the wave function for the coupling matrix in Eq. $^3$.

While Reference $^4$ simulates the spontaneous radiative decay of the excited state by introducing artificial channels, here we include a complex term $-i\hbar \gamma/2$ in each of these excited state potentials to simulate this decay, where in our numerical calculations we take $\gamma = \gamma_m$. This leads to a non-unitary ground state $S$-matrix element such that $0 \leq 1 - |S(k)|^2 \leq 1$ gives probability of atom loss during a collision. In order to avoid problems with spurious asymptotic decay of the field dressed states, a tapered cutoff function is introduced so that decay is only turned on inside some characteristic distance, which we take to be 500 a$_0$. We have verified that $a(k)$ is independent of this choice over the full range of detunings, even between resonances, and $b(k)$ is independent of this choice out to many linewidths away from an isolated resonance. However, in the far wings of a resonance between isolated resonances, $b(k)$ will depend on this choice, although molecular loss tends to be small in these regions. This dependence on cutoff is because our non-unitary theory does not distinguish between atomic and molecular light scattering when the atoms are far apart: the cutoff selects which distance inside of which an excitation is considered to be a molecular loss process instead of an atomic one. Thus the cutoff is taken to be on the order of but larger than the outer turning point of the excited molecular bound states.

Finally, we ignore coupling of the excited $J = 1$ molecular levels back to $d$-waves in the ground state. This means that our model only gets the $s$-wave but not the $d$-wave contributions to the excited state light shifts $^3$. It would be straightforward to add an extra ground state channel to the numerical calculation to account for this.

While there are a number of ways our simple model can be improved, it gives a practical way to calculate the needed complex scattering length for characterizing the intercombination line OFR spectrum of alkaline earth atoms. The model gives two key results. First, it should correctly describe the variation of the real part of the scattering length as laser frequency is tuned across multiple resonances. Second, the isolated resonance approximation should correctly describe the molecular loss rate out to order of hundreds of line widths away from individual isolated resonances.

**Experimental Setup**

Details of the experimental setup can be found in Ref. $^8$. Atoms are loaded into an optical dipole trap...
from a magneto-optical trap operating on the $^1S_0-^3P_1$ intercombination transition. A horizontal (H) and a vertical (V) beam intersect in the $\hat{x} - \hat{z}$ plane to form a crossed dipole trap at the origin shown in Fig. 5 (drawing is to scale). Both beams are derived from the same 1064 nm laser and are linearly polarized along the $\hat{y}$ axis with $1/e^2$ waists 63 µm and 53 µm, respectively. The optical beams point in the directions

\begin{align}
\hat{k}_H &= -\cos \theta_H \hat{x} - \sin \theta_H \hat{z}, \\
\hat{k}_V &= -\cos \theta_V \hat{z} + \sin \theta_V \hat{x}, \\
\hat{k}_{PA} &= -\cos \theta_{PA} \hat{x} + \sin \theta_{PA} \hat{y},
\end{align}

with $\theta_H = 16.0^\circ$, $\theta_V = 14.4^\circ$, and $\theta_{PA} = \pi/8$. The beams are linearly polarized along $\hat{e}_H = \hat{e}_V = \hat{y}$ and $\hat{e}_{PA} = \hat{z}$. The image plane is spanned by $\cos \frac{\pi}{8} \hat{x} - \sin \frac{\pi}{8} \hat{y}$ and $\hat{z}$. A bias magnetic field of $B_z \approx 100$ mGauss defines the atomic quantization axis.

![Diagram showing the geometry of the beam directions and trap fields](image)

**FIG. 5:** Geometry of the experiment in the absorption image plane. The directions $\hat{x}$, $\hat{y}$, and $\hat{z}$ define the lab frame, where both gravity $\hat{g}$ and bias magnetic field $\hat{B}$ are parallel to $\hat{z}$. Symbols $\hat{k}$ are beam directions, and $\hat{e}$ are beam polarization vectors, where subscripts $H$, $V$, and $PA$ indicate horizontal, vertical, and PA beams. $H$ and $V$ Gaussian beam profiles are shown in blue outline, PA Gaussian beam profile in red outline.

The focal condition is not critical to model the potential sufficiently well because of the large Rayleigh ranges ($>8$ mm), and the Gaussian beam isosurfaces at the beam waist are almost cylindrical (blue outlines). Thus, the dipole trap potential due to each beam is modeled as

$$U_i(x) \equiv -U_i^T \exp \left\{ -\frac{2}{w_i^2} \left( [\hat{e}_i \cdot \hat{x}]^2 + [\hat{\kappa}_i \times \hat{e}_i] \cdot \hat{x} \right)^2 \right\},$$

with individual Gaussian beam trap depth [9]

$$U_i^T = \frac{P_i}{\pi \epsilon_0 w_i^2} \text{Re} \alpha_{1S_0}(1064 \text{ nm}),$$

and total laser power $P$. The real part of the dynamic $^1S_0$ polarizability at 1064 nm is $\text{Re} \alpha_{1S_0}(1064 \text{ nm}) \approx 239$ a.u. [10], with atomic unit of polarizability $4\pi\epsilon_0 a_0^3$.

The full model potential includes the gravitational acceleration $g$ pointing along $-\hat{z}$ and is given by

$$U(x) = U_H(x) + U_V(x) + m_S g z.$$  (17)

Gravity sets the trap depths of $\sim 7 \mu K$ and $\sim 15 \mu K$ along V and H. The two graphs on the right hand side of Fig. 5 show cuts through the model potential, which has been adjusted to match the measured trapping frequencies. An isosurface (dark blue) of the model potential at 7 $\mu K$ is shown in the zoomed-out portion.

Typical kinetic energies are 2-4 $\mu K$, and in-trap Full Widths at Half Maximum (FWHM) are 45-55 $\mu m$. The PA beam (red outline) propagates along the horizontal image axis in the $\hat{x} - \hat{y}$ plane with a waist $w_{PA} = 41 \mu m$ and is linearly polarized along $\hat{z}$. Although the PA beam diameter $2\sqrt{2ln2} \times w_{PA} \approx 97 \mu m$ is larger than the typical cloud FWHM, we use a density-averaged intensity $I_{sv} = \int d^3x \rho(x) I(x)/ \int d^3x \rho(x)$ to characterize the PA intensity interacting with the atoms. Typical values are $I_{sv} \approx (0.6 - 0.7) \times I_{pk}$, where $I_{pk} = \frac{2\pi}{\pi w_{PA}^2}$ is the Gaussian beam peak intensity for total beam power $P$.

In addition to scattering from the atomic transition with rate $\Gamma_{sc}$, the PA beam also adds to the optical trap slightly. For large intensities and small $w_{PA}$ and especially in a standing wave configuration, this effect can become important. In this work, the additional trap depth introduced by the PA beam is typically $< 0.1 \mu K$.

**Imaging Procedure and Analysis**

We use a two-component long-working-distance microscope with 3 $\mu m$ resolution at 532 nm. The imaging lens is placed outside the vacuum chamber at a distance of 150 mm from the atomic cloud. The second part of the telescope is an eyepiece mounted to a CCD camera with $1024 \times 1024$ square pixels of 13 $\mu m$ width. The eyepiece includes an interference filter at 461 nm (intensity transmission 0.45 within 10 nm bandwidth, ND 5.0 otherwise). Time of flight measurements were used to determine an imaging magnification of 6.5, corresponding to 2.0 $\mu m$ per pixel. We tested the imaging system resolution in a separate test setup that included the effect of an anti-reflection (AR) coated vacuum viewport, same as the one used on our vacuum chamber. Using incoherent white light filtered by the blue interference filter with transmission (AR) coated vacuum viewport, same as the one used on our vacuum chamber. Using incoherent white light filtered by the blue interference filter with transmissive test targets, resolutions of $\sim 4 \mu m$ were measured. Absorption imaging with coherent light causes etaloning between the CCD chip and the glass plate that covers it. To reduce fringing, the glass plate surface is wedged at $\sim 1^\circ$ and AR coated for the imaging wavelength.

Without the elastic contribution of the OFR effect, the $^{88 Sr}$ sample does not thermalize on experimental timescales due to its small $a_{og}$, and it is important to obtain information about both the potential and kinetic...
energies of the gas. We interleave experimental runs between in-situ imaging of the sample and imaging after the atoms were dropped for 1.5 ms by switching off the optical dipole trap. The first image gives information about the spatial density of the atomic cloud, and the second image measures the kinetic energy by time-of-flight expansion. We typically repeat this interleaved sequence 3-5 times and average the resulting in-situ and time-of-flight absorption images before performing data analysis. We also calculate the pixel-by-pixel studentized standard deviations which are used as weights in fitting the mean image and also enter the fit parameter error estimates. Both the mean and standard-deviation images are then rotated (with bilinear interpolation) by 14° about the center of the atom cloud to transform the images into the eigenframe of the trap. The pictures are then integrated along the horizontal (vertical) axis and are fit to a one-dimensional Gaussian distribution including a background offset and linear slope to account for residual fringing and CCD readout noise in the absorption image.

A common problem in imaging atomic clouds with large optical depth is light that does not interact with atoms hitting the camera in the same position. Common causes are forward scattering of photons into the imaging path or mixed probe polarization. Probing the \( ^1\text{S}_0-^1\text{P}_1 \) transition is insensitive to probe polarization even under a small bias magnetic field. To identify the possibility of a limiting optical depth \( \text{OD}_{\text{ceiling}} \), a series of images were taken with increasing atomic density. We compared the peak optical depth in-situ versus the pixel-summed optical depth either of an in-situ or a corresponding time-of-flight image, and we estimate a conservative limit \( \text{OD}_{\text{ceiling}} \geq 3.5 \). Typical in-situ optical depths in this paper were below 2, so that we can neglect the effect.

For probe intensities larger than 0.1\( I_{\text{sat}} \), with saturation intensity \( I_{\text{sat}} \approx 40 \text{ mW/cm}^2 \), saturation of the imaging transition becomes important. To account for the saturated absorption, we correct the full expression for the OD to the linear Beer-Lambert regime \( \text{OD} \). For each picture the saturation correction is applied on a pixel-by-pixel basis.

While the shadow image is forming and the atoms are scattering light, their momentum undergoes a random walk due to the spontaneous emission of blue photons. By examining the shadow image versus a reference image without atoms, we extract the number of photons that the atoms removed from the probe pulse. By varying the probe time, we find the cloud expansion as a function of the number of photons scattered and apply a corresponding correction to the cloud widths from both in-situ images and TOF images.

We extract the integrated OD of the mean image to obtain the atom number. Corrections for probe heating are then applied to the fitted widths. The in-situ image widths are used to calculate the mean atomic density. Finally, we calculate a time-of-flight temperature corrected for finite size effects by the in-situ widths.

**Loss Spectra** \( 2k\ell_{\text{opt}}\gamma_m/\gamma \ll 1 \)

To extract \( \ell_{\text{opt}}\gamma_m \) from the loss spectra, data was fit to an approximate expression for the integral of the atomic density, where the density after the PA pulse was calculated via the differential equation \( \dot{\rho} = -K_{\text{in}}(\rho - \rho_{\text{bg}}) \). The initial density distribution was modelled as Gaussian with \( w_H \) and \( w_V \). The width into the plane of the images was extrapolated from a Monte-Carlo simulation of the trap model potential. The fitting function is

\[
N = \frac{2}{\sqrt{\pi}} \int_0^\infty du \frac{\sqrt{u} e^{-u}}{1 + K_{\text{in}}(\Delta, u, \ell_{\text{opt}}\gamma_m, T) \tau_{\text{eff}} \rho_0 e^{-u}},
\]

with normalized dimensionless trap length scale \( u \). This expression describes the number signal normalized to the measured atom number after the PA pulse of duration \( \tau_{\text{PA}} \) and when the detuning from molecular resonance is large. The quantity \( \tau_{\text{eff}} = \tau_{\text{bg}} e^{\gamma \text{in} \ell_{\text{opt}}\gamma_m \ell_{\text{eff}} / \tau_{\text{bg}} \gamma_{\text{in}} \ell_{\text{opt}} \gamma_m - 1} \) accounts for the fact that atoms are lost to one-body decay during the PA pulse with lifetime \( \tau_{\text{bg}} \) and that we hold the atoms for \( \tau_{\text{hold}} = 100 \text{ ms} \) between the end of the PA pulse and the imaging. The number \( \rho_0 \) is the peak density after the PA pulse and at large detuning.

The definition of all other symbols follows the main text. To make the fit numerically tractable, the integral was approximated as a ten-term sum using Gauss-Laguerre quadrature.

The thermally-averaged inelastic collision rate per particle \( K_{\text{in}} \) was derived from the s-wave scattering matrix of Bohm and Julienne, using the definition of the optical length \( \ell_{\text{opt}} = \Gamma_s / 2k\gamma_m \) in terms of the stimulated width \( \Gamma_s \). Here, we use a thermally-averaged inelastic rate coefficient \( \bar{K}_{\text{in}} = \langle \hbar k \sigma_{\text{in}} / \mu \rangle \), where \( \sigma_{\text{in}} = 2 \hbar^2 (1 - |S|^2) \). The angular brackets denote a thermal average over the initial collision momenta. This average was performed on the inelastic rate coefficient (rather than the density after the PA pulse) under the assumption that prior to the PA pulse, many single-particle velocities exist in every differential volume in the trap. Since single collisions take place in a small volume, the spatial dependence of \( K_{\text{in}} \) was carried through into the differential equation for the density rather than averaged out.

To fit the inelastic loss spectra, \( \bar{K}_{\text{in}} \) was expressed as

\[
\bar{K}_{\text{in}}(\Delta, u, \ell_{\text{opt}}\gamma_m, T) = \frac{8\sqrt{\pi} \hbar \ell_{\text{opt}}\gamma_m}{\mu} \int_0^\infty d\eta \frac{\gamma \sqrt{\eta} e^{-\eta}}{D^2 + 1/4},
\]

\[
D \equiv \Delta + \frac{k_B T}{\hbar} \eta - \nu_{\text{rec}} - \nu_{\text{th}} e^{-u/u_0},
\]

\[
\Gamma \equiv \gamma + 2k_{\text{th}} \ell_{\text{opt}}\gamma_m \sqrt{\eta},
\]

(19)
with dimensionless relative momentum magnitude \( \eta \equiv k^2/\ell_{\text{opt}}^2 \), thermal momentum \( \hbar k_{\text{th}} = \sqrt{2m k_{\text{th}}^2 T} \), center-of-mass PA photon recoil energy \( h\nu_{\text{rec}} \), trapping laser ac Stark shift at the center of the trap \( \nu_s \), and Planck's constant \( h = 2\pi\hbar \). The PA laser with optical frequency \( \nu_l \) is detuned from the PA resonance by
\[
\Delta \equiv \nu_l - [\nu(1S_0 - 3P_1) + \nu_n + \Delta\nu(I)],
\]
where \( \nu(1S_0 - 3P_1) \) is the atomic transition frequency, and \( h\nu_n \) is the energy of state \( n \) with respect to the free threshold. The detuning term \( \Delta\nu(I) \) accounts for the ac Stark shift of the molecular resonance with respect to \( I \).

The integral in Eq. (19) was approximated as a 53-term sum using Gauss-Laguerre quadrature. The quantities \((\ell_{\text{opt}}\gamma_m), T, u_0, \) and a term added to the detuning to represent the line center were allowed to vary. The parameter \( T \) is used as a check against the experimentally measured temperatures and agrees well with the experiment. The Stark shift term \( \nu_s e^{-\eta/N_0} \) was included to account for the broadening of the atomic loss profile to the blue side of a molecular resonance due to the ac Stark shift induced by the trap. The molecular line width \( \gamma \) was extracted from the Monte-Carlo simulation in the next section.

### Monte-Carlo Simulation (2k\( \ell_{\text{opt}}\gamma_m/\gamma \sim 1 \))

To model the thermodynamic effects caused by the interplay of elastic and inelastic collisions in an anharmonic trap with evaporation, we use a Monte-Carlo simulation since analytic expressions such as those presented in the previous section are not available. The simulation is based on classical particles moving in a conservative model potential that includes the Gaussian beam shapes as well as gravity. A commonly used method \[\[13, \[14, \[15\] to include collisions in such simulations is due to Bird \[16\] and detailed discussions of the method in the context of ultracold atoms in optical traps can be found in Refs. \[17, \[18\]. The simulation procedure here is described in more detail in Ref. \[8\].

The simulation uses the elastic and inelastic cross section formulas derived in Section “Scattering Matrix, Cross Sections, and Collision Rates”:
\[
\sigma_{\text{el}}(k) = \frac{4\pi \ell_{\text{opt}}^2 \gamma_m}{k (\Delta + E/h)^2/\gamma^2 + (1 + 2k\ell_{\text{opt}}^2 \gamma_m)^2/4},
\]
where the second relation is an approximation valid in an ideal ultracold gas like \(^{88}\)Sr where the background scattering length \( a_{\text{bg}} \) can be neglected. For a gas at nonzero temperature, the detuning \( \Delta \) includes the atomic motion and the trap ac Stark shift as in the previous section.

The initial particle distribution is synthesized by dropping atoms into the model potential and letting them evolve for several hundred ms without collisions (using an embedded Runge-Kutta method). Each particle is initially generated from independent Gaussian distributions along the trap eigenaxes, both in position and velocity. The initial widths of these Gaussian distributions are adjusted until the particle distribution after having settled in the model potential matches the experimental in-situ and TOF data when the PA laser is far detuned from the OFR resonance.

To calibrate the Monte-Carlo simulation, we modelled a three-dimensional isotropic harmonic trap of \( \mu K \) trap depth and checked the thermodynamic effects of inelastic and elastic collisions independently. Using a Gaussian initial phase-space density at several \( \mu K \) and only allowing inelastic collisions at a velocity-independent cross section, we recovered heating rates per particle that match the corresponding analytic expressions \[8\]. Similarly, we check cross-dimensional thermalization rates at a velocity-independent elastic cross section. We found that the average number of elastic collisions required for cross-dimensional thermalization in our simulation agrees \[8\] with the analytic expressions from Ref. \[15\].

The Monte-Carlo simulation also includes the effect of scattering from the atomic line. For the largest optical lengths in Fig. 4 of the main paper, the atomic scattering rate is \( \sim 6 \text{s}^{-1} \) corresponding to 1.2 photons scattered per atom during the 200 ms exposure time. At these scattering rates, the photon absorption along the PA laser direction and the random reemission change the resulting cloud widths by less than 10\%. Due to the spontaneous reemission as a spherical wave, the mean free path of a scattered photon in a sample of OD \( \sim 2 \) is still larger than the typical cloud radii and we estimate that radiation trapping should only introduce a small correction.

The final particle distribution is then imaged by randomly scattering photons off of each particle and forming a histogram from the positions of the scattering events over the image pulse time of 50 \( \mu \text{s} \), either in the trap or after TOF expansion for 1.5 ms. Random rescattering and corresponding velocity and position evolution are included, and the position histogram is blurred to account for the 4 \( \mu \text{m} \) image resolution. The final image is then analyzed in the same way as the experimental data to extract in-trap widths and TOF temperatures. By adding the final measurement step instead of calculating the covariance matrices of the position and velocity distributions directly, the quantitative agreement with the experiment was improved significantly in the regime of large inelastic losses.
Maximal scattering length modification

In the following, we build on the understanding from our current experiment and try to estimate the maximum scattering length under the most ideal conditions. We assume that atomic scattering is the dominant loss mechanism, that $k \to 0$, and that there is no extra molecular loss, such that $\gamma = 2\gamma_a$. There is also no spatial inhomogeneity, and thus it does not reflect the current experimental conditions. Theoretical calculations of the molecular line strength factors are used to make these estimates. Also note that the line strength factors are only order-of-magnitude estimates beyond $n < -5$.

The Optically-Modified Scattering Length Constrained by Atomic Light Scatter

In the $k \to 0$ limit, the scattering length change $\Delta a$ due to an optical Feshbach resonance is given by

$$\Delta a = \ell_{\text{opt}} \frac{(\delta - \delta_0)\gamma_m}{(\delta - \delta_0)^2 + \gamma_m^2/4} = 2\ell_{\text{opt}} \frac{(\delta - \delta_0)\gamma_a}{(\delta - \delta_0)^2 + \gamma_a^2}, \quad (22)$$

where $\delta$ is $2\pi$ times the detuning from atomic resonance, $\delta_0$ is the difference between the molecular and atomic resonance frequencies, $\gamma_m$ is the molecular linewidth, $\gamma_a$ is the atomic linewidth, and $\gamma_m = 2\gamma_a$. The quantity $\ell_{\text{opt}} = \xi I$, where $I$ is the PA laser intensity and $\xi \equiv \frac{I}{\lambda^2/((\pi k)^2) I}$ is a resonance-specific constant that describes the Sr+Sr molecular structure.

When using an OFR, $\delta$ must be chosen such that inelastic collisional loss is small. In this regime, loss due to elastic collisional loss is small. In this regime, loss due to atomic light scattering dominates. The atomic scattering rate $\Gamma_{\text{sc}}$ is given by

$$\Gamma_{\text{sc}} = \frac{\gamma_a}{2} \frac{s_0}{1 + s_0 + 4(\delta/\gamma_a)^2}. \quad (23)$$

Here $s_0 = I/I_{\text{sat}}$, where $I_{\text{sat}}$ is the saturation intensity of the atomic transition. Typically the experiment determines the maximum allowable value of $\Gamma_{\text{sc}}$, thereby constraining $I$. Under this constraint, $I$ can be expressed as

$$I = I_{\text{sat}} \frac{2\Gamma_{\text{sc}}/\gamma_a}{1 - 2\Gamma_{\text{sc}}/\gamma_a} \left(1 + 4\frac{\delta^2}{\gamma_a^2}\right) \approx 8I_{\text{sat}} \left(\frac{\Gamma_{\text{sc}}}{\gamma_a}\right) \left(\frac{\delta}{\gamma_a}\right)^2, \quad (24)$$

where it has been assumed that $\gamma_a \gg \Gamma_{\text{sc}}$ and $\delta \gg \gamma_a$, the latter of which is true due to strontium’s narrow intercombination line that is used in our work. The maximum $\ell_{\text{opt}}$ for a given detuning is simply

$$\ell_{\text{opt}} = \xi I = 8\xi I_{\text{sat}} \left(\frac{\Gamma_{\text{sc}}}{\gamma_a}\right) \left(\frac{\delta}{\gamma_a}\right)^2. \quad (25)$$

Thus $\Delta a$, constrained to a certain $\Gamma_{\text{sc}}$, is

$$\Delta a = 16\xi I_{\text{sat}} \left(\frac{\Gamma_{\text{sc}}}{\gamma_a}\right) \frac{\delta^2}{\gamma_a^2} \left(\frac{\delta - \delta_0}{}\right) \left(\frac{\gamma_a}{\gamma_a^2 + \gamma_a^2}\right). \quad (26)$$

This expression is plotted in Figs. 6 and 7. Within tens of linewidths $\gamma_m$ of $\delta = \delta_0$, the factor $\delta^2$ in the numerator of Eq. 26 is slowly varying and can be approximated as $\delta_0^2$. Let $\Delta a_{\text{dis}}$ refer to $\Delta a$ in this regime. Therefore,

$$\Delta a_{\text{dis}} = 16\xi I_{\text{sat}} \left(\frac{\Gamma_{\text{sc}}}{\gamma_a}\right) \frac{\delta_0^2}{\gamma_a^2} \left(\frac{\delta - \delta_0}{}\right) \left(\frac{\gamma_a}{\gamma_a^2 + \gamma_a^2}\right), \quad (27)$$

which has the dispersion shape of Fig. 6. When $\delta \gg \delta_0$, $|\Delta a|$ varies linearly with $\delta$. Let $\Delta a_{\text{lin}}$ be the expression...
for $\Delta a$ when $\delta \gg \delta_0$.

$$\Delta a_{\text{lin}} = 16\xi I_{\text{sat}} \left( \frac{\Gamma_{sc}}{\gamma_a} \right) \left( \frac{\delta}{\gamma_a} \right),$$

(28)

which describes the linear behavior evident in Fig. 4.

The Maximum Useful Scattering Length Near a Molecular Resonance

![Graph showing the maximum useful scattering length near a molecular resonance](image)

FIG. 8: Equation 26 plotted in blue and Eq. 29 plotted in red. This plot was generated with the same values for $\xi$, $\delta_0$, $\Gamma_{sc}$, $\gamma_a$, and $I_{\text{sat}}$ as those used in Fig. 4. Note that $K_{in}$ is significant near a molecular resonance but that this quantity drops off faster than $\Delta a$.

Near a molecular resonance where $|\Delta a|$ has dispersive behavior, it follows from Eq. 27 that $|\Delta a|$ has a local maximum at $|\delta - \delta_0| \approx \pm \gamma_a$. However, at these small detunings inelastic collisional loss must be considered. For $k \to 0$, the inelastic rate coefficient $K_{in}$ is given by

$$K_{in} = \frac{4\pi \hbar_{\text{opt}}}{\mu} \left( \frac{\gamma_m}{\delta - \delta_0} \right)^2 + \frac{\gamma_m^2}{4},$$

(29)

Putting Eq. 29 into Eq. 26 it can easily be shown that the maximum value of $K_{in}$, denoted by $K_{in}^{\text{max}}$, occurs at $\delta \approx \delta_0$, consistent with the case when $\ell_{\text{opt}}$ in Eq. 26 is not constrained by atomic light scatter.

As Fig. 5 shows, inelastic loss (described by $K_{in}$) is significant near $\delta_0$. Nevertheless, this figure makes it apparent that one can choose a detuning that is just outside of the influence of $K_{in}$ but that still yields a significant $\Delta a$. This detuning is implied upon setting $K_{in} = \kappa K_{in}^{\text{max}}$, where $\kappa$ describes the fraction of molecular loss deemed acceptable by the experiment. Solving this equation for detuning yields that $\delta - \delta_0 \approx \pm \gamma_a / \sqrt{\kappa}$. Evaluating Eq. 26 at this detuning produces $\Delta a_{\text{max}}$,

$$\Delta a_{\text{max}} \approx \pm 16\xi I_{\text{sat}} \sqrt{\kappa} \left( \frac{\Gamma_{sc}}{\gamma_a} \right) \left( \frac{\delta_0}{\gamma_a} \right)^2,$$

(30)

where $\Delta a_{\text{max}}$ is the maximum scattering length change when $\Delta a$ is dispersive and inelastic loss and atomic light scatter are negligible. The intensity corresponding to $\Delta a_{\text{max}}$, given by Eq. 24 is

$$I_{\Delta a_{\text{max}}} \approx 8I_{\text{sat}} \left( \frac{\Gamma_{sc}}{\gamma_a} \right) \left( \frac{\delta_0}{\gamma_a} \right)^2.$$  

(31)

The Large-Detuning Case

Although $|\Delta a|$ in Eq. 26 seems to increase without bound when $\delta \gg \delta_0$, coupled-channels OFR theory (which so far has not been considered in this discussion) dictates that if $|\Delta a|$ is based on the $n$th resonance, the modification to the length will vanish at some point before $\delta$ is equal to the detuning of the $(n - 1)$th resonance. See the main text for a further discussion of this coupled-channels effect. One must consider whether the molecular detuning $\delta - \delta_0$ can be sufficiently large to take advantage of the linear increase with $\delta$ but not so far that coupled-channels effects become a concern.

Let $\delta_{lin}$ be the detuning at which the magnitude of $\Delta a_{\text{lin}}$ is equal to $|\Delta a_{\text{max}}|$. It follows that $|\delta_{lin}| = \sqrt{\kappa \delta_0^2 / \gamma_a}$. If $\delta_{lin}$ is comfortably outside the regime where coupled-channels effects are a concern, then the maximum useful scattering length occurs when one is many linewidths $\gamma_m$ detuned from $\delta_0$ but not far enough detuned to be close to other resonances. However, theoretical values for $\delta_0$ make it clear that detuning to $\delta_{lin}$ would always require crossing another molecular resonance, so coupled-channels effects dictate that the maximum scattering length one can achieve is given by $\Delta a_{\text{max}}$ (Eq. 26), occurring when $\delta$ is as close as possible to a molecular line, while molecular losses are constrained to a given level. Table 1 considers 6 different molecular resonances and their associated values of $\delta_{lin}$, which are many orders of magnitude larger than $\delta_0$ for subsequent resonances.

| $n$ | $\delta_0$ (GHz) | $\delta_{lin}$ (GHz) |
|-----|-----------------|---------------------|
| -2  | -0.023          | -7.22               |
| -3  | -0.221          | -655                |
| -4  | -1.084          | -1.57 x 10^4        |
| -5  | -3.460          | -1.60 x 10^5        |
| -6  | -8.400          | -9.40 x 10^5        |
| -7  | -17.0           | -3.84 x 10^6        |

TABLE I: Theoretical values for $\delta_0$ for the $n = -2$ through -7 $\theta_a$ resonances and the associated values for $\delta_{lin}$. Here $\kappa = 0.01$. 


According to Eq. 30, only moderate gains in $\Delta \delta$ will only be accompanied by a $\sqrt{\kappa}$ increase in $\frac{\Gamma_{\text{sat}}}{I}$ since any increase in $K_{\text{in}}$ will only be accompanied by a $\sqrt{\kappa_{\text{in}}}$ increase in $|\Delta \theta_{\text{max}}|$. More significant gains can be achieved by shortening experimental time scales to allow for larger values of $\Gamma_{\text{sc}}$ since $|\Delta \theta_{\text{max}}|$ is linear in $\Gamma_{\text{sc}}$.

TABLE II: Theoretical values for $\delta_0$ and $\xi$ for the $n = -2$ through -7 $0_u$ resonances are used to calculate the maximum useful scattering length. The intensities required for the se

$$
\begin{array}{|c|c|c|c|}
\hline
n & \delta_0 \text{ (GHz)} & \xi \text{ (} a_0/(W/cm^2)\) & |\Delta \theta_{\text{max}}| \text{ (} a_0 \) & I_{\Delta \theta_{\text{max}}} \text{ (W/cm}^2) \\
\hline
-2 & -0.023 & 6110 & 5.97 & 4.89 \times 10^{-3} \\
-3 & -0.221 & 32.8 & 2.92 & 0.443 \\
-4 & -1.084 & 27.9 & 59.3 & 10.6 \\
-5 & -3.460 & 0.272 & 71.5 & 108 \\
-6 & -8.400 & 0.272 & 34.7 & 637 \\
-7 & -17.0 & 0.012 & 6.21 & 2.61 \times 10^3 \\
\hline
\end{array}
$$

Conclusion

In light of Section “The Large-Detuning Case”, we conclude that for a given molecular line, the best optically-modified scattering length occurs for a laser detuned just outside the influence of inelastic loss and with an intensity just low enough to prevent heating due to atomic light scatter. This optimal detuning is given by $\delta - \delta_0 = \gamma_a / \sqrt{\kappa}$. Table II lists theoretical $0_u$ values for $\xi$ and $\delta_0$ as well the associated values for $\Delta \theta_{\text{max}}$ and $I_{\Delta \theta_{\text{max}}}$. Values of $\Gamma_{\text{sc}} = 1 \text{ s}^{-1}$ and $\kappa = 0.01$ have been used. For the strontium $^1S_{0,3}P_1$ line, $\gamma_a = 2\pi \times 7.5$ kHz and $I_{\text{sat}} = 3 \mu \text{W/cm}^2$. These numbers yield an optimal detuning of $\delta - \delta_0 = 2\pi \times 75$ kHz. The intensity column was included to point out that many resonances cannot be used on technical grounds (for instance intensities greater than a few kW/cm$^2$ are impractical for external cavity diode lasers).

Under the ideal conditions of zero temperature, no spatial inhomogeneity, no extra molecular loss and the assumption that atomic light scattering is the dominant loss mechanism, Table II shows that the resonance location of the $n = -5$ line is the best compromise between a large detuning from atomic resonance (which allows for more PA laser intensity without atomic light scatter) and the fact that $\xi$ rapidly decreases with increasing $|\delta|$. According to Eq. 30 only moderate gains in $\Delta \theta_{\text{max}}$ can be achieved by allowing for inelastic loss (when detuning closer to a molecular line) since any increase in $K_{\text{in}}$ will only be accompanied by a $\sqrt{\kappa_{\text{in}}}$ increase in $|\Delta \theta_{\text{max}}|$.

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