Prospects for influencing scattering lengths with far-off-resonant light

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We explore a recent proposal [Fedichev *et al.*, Phys. Rev. Lett. **77**, 2913 (1996)] for altering the mean interaction strength between ultracold atoms using an appropriately detuned laser. Although care must be taken to minimize laser-driven loss processes, we find large ranges of intensities and detunings where useful changes might be affected. Accordingly, we present simple formulas for the effects of laser light that should prove useful in designing specific experiments. We demonstrate the validity of these formulas by comparison with exact close-coupling models. In particular, we find that useful changes of the mean-field interaction require sufficiently high laser intensities that the rate of laser-induced *stimulated* emission exceeds the natural *spontaneous* emission rate. [S1050-2947(97)08308-X]

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I. INTRODUCTION

The emerging science of Bose-Einstein condensation (BEC) in dilute samples of trapped atomic gasses demands new tools for studying and probing this unique state of matter. Atoms in condensates collectively experience distortions and shifts in energy levels beyond those imposed by their harmonic trap environments, as dictated by their weak interatomic interactions. The influence of these interactions can be adjusted macroscopically by varying the density of the entire cloud, for instance, by varying the trap's confining potentials or the number of trapped atoms [1]. In addition, the spectroscopy performed so far on condensates has relied on jiggling the magnetic trapping potential at a given frequency and gauging the amplitude of the cloud's response [2]. The resulting distortions and spectra of the condensed clouds have been adequately addressed within a mean-field picture, emphasizing the average motion of each atom against the background of its counterparts [3].

A more microscopic probe of cold atom clouds would directly tune the interatomic interactions, encapsulated at extremely low temperatures in the s-wave scattering length a. The mean-field interaction strength over the whole cloud is transmitted through elastic collisions, whose influence in turn is described by the zero-energy elastic cross section for identical bosons, $\sigma_{\rm el}=8\pi a^2$. The sign of a also proves relevant to BEC physics, since condensates with negative scattering lengths (implying an effectively attractive interaction between atoms) are understood to be stable only up to a certain population of atoms [4]. In addition, a ''double condensate'' of two distinct spin states of ^{87}Rb has recently been achieved [5], implying three different scattering lengths between different pairs of collision partners. Altering one or more of these scattering lengths independently of the others suggests a rich reaction physics in such clouds.

The prototypical scheme for altering scattering lengths suggests applying a magnetic field to alkali-metal atoms trapped in their $|FM_F\rangle=|1,-1\rangle$ hyperfine state. [6]. A magnetic field of the appropriate strength then induces spin-flip transitions of one or both atoms into higher-energy $|FM_F\rangle=|2,2\rangle$ hyperfine states, from which the atoms have insufficient energy to escape from one another. The resulting quasibound state forms a resonance in which the relative motion of the atom pair can be severely altered, thus affecting the effective scattering length. Such resonances now appear to be accessible in certain species, for instance ^{85}Rb, but not in others, such as ^{87}Rb [7].

An alternative possibility applicable to any species employs a laser tuned near resonance with an excited molecular level, in the scheme used for photoassociation (PA) spectroscopy [8]. Here the excited state potential takes the form $-C_3/R^3$ in the ranges of internuclear separation R of interest. The resulting large acceleration of the atoms toward one another can strongly affect scattering lengths even far away from a vibrational resonance in the excited state potential, as suggested by Fedichev *et al.* [9]. The drawback to this method for influencing scattering lengths is that, near a PA resonance, the quasibound molecular state suffers spontaneous emission events which almost invariably lead to trap loss. In addition, individual atoms can scatter laser photons inelastically, picking up sufficient energy to leave the trap. Nevertheless, Ref. [9] asserted that brief pulses of laser light can significantly alter interatomic scattering lengths without ejecting too many atoms from the trap. Both loss processes can be reduced by detuning away from resonances, but doing so requires higher intensities to achieve a desired change of scattering length.

To quantify these effects, this paper provides a more detailed account of the process described in Ref. [9] by presenting the results of close-coupling calculations for atomic scattering near threshold in a laser field. We will demonstrate the general usefulness of a simple formula, involving only the Franck-Condon factors for the relevant transitions, that gives good agreement with the close-coupled equations for changes in scattering lengths and for two-body trap loss

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rates. This paper is thus intended as a guide to experimental efforts intent on making a sudden, large change in scattering length over a limited period of time.

Such a sudden change might prove useful in laser-assisted evaporative cooling of atomic clouds, where atoms are routinely ejected from the cloud anyway. Another use would be as a spectroscopic probe that examines the cloud's response when kicked by the laser at intervals near the period of the cloud's natural oscillation. Moreover, a probe that changes only the scattering length (and not the overall density or trap spring constants) might be able to probe spectroscopically the dense condensate portion of the cloud, independently of the background of thermal atoms in which it swims. This kind of probe might help in sorting out the surprising measurements of excitation lifetimes obtained in Bose condensates at nonzero temperatures [10].

II. GENERAL CONSIDERATIONS

Any alteration of the effective interaction between colliding cold atoms must be quick to be effective. In particular, the change in elastic collision properties must take place on a time scale short compared to that of inelastic collisions, which generally produce untrapped spin states and destroy the atom cloud. This requirement establishes an upper limit on the inelastic rate coefficients $K_{\rm inel}$. The allowed upper limit can be different, depending on whether the atom cloud to be altered is a condensate or a thermal cloud.

A condensate's characteristic time scale is set by its excitation frequency $\nu_{\rm cond}$ (typically $\sim$100 Hz in contemporary traps). Measuring the altered excitation frequency, for instance, requires observation of at least several oscilations of the cloud, of duration $1/\nu_{\rm cond}$. During this time, only a small fraction of the atoms can be ejected from the condensate, or else its character will change during the measurment. For a given number density of atoms n, we therefore require

$$nK_{\rm inel} \ll \nu_{\rm cond}. \tag{1}$$

For typical trap densities of $n\sim 10^{14}/{\rm cm}^3$, this criterion requires that, for a useful, sustained change of scattering length, $K_{\rm inel}$ must generally lie well below 10^{-12} cm^3/s. An intended *sudden* change of scattering length of course poses a less serious restriction.

To assist in evaporative cooling of a thermal cloud requires instead that the elastic collision rates should greatly exceed the lossy inelastic rates; that is, the cloud should have adequate time to rethermalize before all the atoms are lost. For the laser-assisted PA process this implies that the elastic collision rate coefficient $K_{\rm el}$ should greatly exceed the inelastic rate coefficient $K_{\rm inel}$ arising from spontaneous emission of the excited molecular state. These rate coefficients are given by the thermal averages

$$K_{\rm el,\ inel} = \langle v_i \sigma_{\rm el,\ inel}(E) \rangle, \tag{2}$$

where $v_i = \hbar k_i/\mu$ stands for the relative incident velocity of the colliding atoms, related to their relative kinetic energy E and reduced mass μ through $E = \hbar^2 k_i^2/2\mu$. $\sigma_{\rm el,\ inel}(E)$ represents the scattering cross section for elastic and inelastic processes, respectively. For the sub-mK temperatures relevant in these experiments, the scattering processes lie within the Wigner threshold regime, so that s-wave collisions will dominate. In addition, the average in Eq. (1) reduces at these temperatures to evaluating $v_i\sigma(E)$ at a single-scattering energy $E = k_B T$ corresponding to the trap temperature T.

In the threshold regime, we account for inelastic processes by introducing an imaginary part to the phase shift, following Mott and Massey [11]:

$$\delta = \lambda + i\mu, \quad \mu > 0. \tag{3}$$

This complex pahse shift leads to a scattering matrix element of the form

$$S = \exp[2i(\lambda + i\mu)]. \tag{4}$$

The magnitude of S is less than unity, accounting for flux lost to the spontaneous emission channel. In the low-energy limit, the complex phase shift translates into a complex scattering length

$$a = A - iB \equiv -\lim_{k_i \to 0} \frac{1}{k_i} \tan(\lambda + i\mu). \tag{5}$$

The threshold scattering cross sections can then be written in terms of A and B as

$$\sigma_{\rm el} \approx 8\pi(A^2 + B^2), \quad \sigma_{\rm inel} \approx 8\pi \frac{B}{k_i}. \tag{6}$$

These cross sections yield in turn the rate coefficients

$$K_{\rm el} = \frac{8\pi\hbar}{\mu} k_i (A^2 + B^2), \quad K_{\rm inel} = \frac{8\pi\hbar}{\mu} B, \tag{7}$$

where each inelstic scattering event accounts for the loss of two atoms from the trap. Note that the inelastic rate remains constant as the temperature (and hence k_i) goes to zero, but that the elastic rate *vanishes* in this limit. Therefore maintaining $K_{\rm el} \gg K_{\rm inel}$, as required to make the change of scattering length useful in thermal clouds, becomes increasingly difficult at lower trap temperatures. From Eq. (7), the ''usefulness condition'' $K_{\rm el} \gg K_{\rm inel}$ can be expressed in terms of the reduced de Broglie wavelength $\lambda\!\!\!^{-} = 1/k_i$ of the colliding atoms,

$$A^2 + B^2 \gg B\lambda\!\!\!^{-}. \tag{8}$$

Note that for alkali-metal atoms A and B are typically tens or hundreds of atomic units, while in μK traps the reduced deBroglie wavelengths can reach into the thousands or tens of thousands of atomic units. Equation (8) thus places a very severe limit on the usefulness of this procedure, given that the overall loss rates should remain as small as possible. In particular, this limit is much more restrictive than the criterion $A \gg B$ suggested in Ref. [9].

Finally, we must also be aware of loss due to the scattering of individual atoms by the laser light, whose rate is given by [12]

$$\gamma_{\rm atom} = \gamma_A \left(\frac{\Omega_A}{\Delta_A} \right)^2. \tag{9}$$

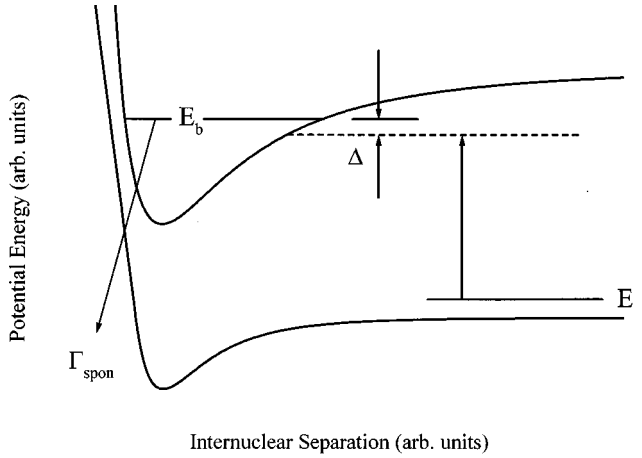

FIG. 1. Schematic plot of the ground- and excited-state molecular potentials. The photoassociation laser is detuned Δ_A to the red of the atomic resonance, and Δ with respect to a vibrational level of the excited potential. This excited state can then decay by spontaneous emission, with a rate $\Gamma_{\rm spon}$.

Here $\Omega_A=(2\pi I/c)^{1/2}d_A$ stands for the atomic Rabi frequency for the $s\rightarrow p$ transition, given an applied laser intensity I and an atomic transition dipole moment d_A; Δ_A stands for the laser's detuning from the atomic resonance. To avoid these one-body loss processes, we should therefore aim at molecular bound levels far detuned from atomic resonance. For typical trap densities ($\sim 10^{13}$–10^{14} atoms cm^{-3}) the product of $K_{\rm inel}$ times atomic density will exceed $\gamma_{\rm atom}$ in the results below, so that we focus our attention primarily on the limitations imposed by two-body losses.

III. MODEL AND SIMPLIFIED FORMULAS

In the present treatment we focus on clouds of atoms trapped in the spin-polarized $|FM_F\rangle=|22\rangle$ hyperfine states, as has been the case in ultracold experiments performed at JILA and at Rice. Pairs of such atoms collide with total electronic spin states of a purely triplet character, which simplifies the analysis below. Collisions between atoms in trapped states other than $|22\rangle$ will require explicit handling of singlet electronic character of the electron spins, as well as the possibility of spin exchange processes that can change the hyperfine states [13].

We therefore view the process at hand as a three-channel scattering problem, depicted schematically in Fig. 1 as a set of molecular potential curves. The incident channel, denoted g, represents two atoms approaching one another with relative kinetic energy E in the ground-state triplet potential. The excited channel e contains the bound state of energy E_b near which the laser of frequency ω is detuned, with detuning $\Delta\equiv E_b-\hbar\omega$. The third channel p represents the product generated by spontaneous emission. In our close-coupling calculations we treat this channel with an artificial channel potential (not shown in Fig. 1) that serves merely as a destination for atoms lost to spontaneous emission.

The radiative coupling between channels g and e in our model takes the R-independent form $(2\pi I/c)^{1/2}d_M$, where I again stands for the laser's intensity and c the speed of light, and now d_M denotes a molecular dipole transition moment, related to the atomic dipole transition moment d_A by $d_M=d_A\sqrt{2}/\sqrt{3}$ for the transitions of interest here [14]. The artificial channel is coupled to the e channel with a strength sufficient to reproduce the natural linewidth for spontaneous emission, in the limit of vanishing laser intensity. The close-coupled calculation first rearranges the potentials into the ''field-dressed'' basis, by diagonalizing the radiative part of the Hamiltonian in the $R\rightarrow\infty$ limit.

As noted above, realistic modification of scattering lengths requires detuning in the vicinity of PA resonances, but not too near. For the present model, the results of full close-coupling calculations agree well with the following semianalytic formulas, which arise from treating the laser field as a perturbation [15]:

$$A=A_0+\frac{1}{k_i}\frac{\frac{1}{2}\Gamma_{\rm stim}[E-\Delta-E(I)]}{[E-\Delta-E(I)]^2+(\Gamma_{\rm spon}/2)^2-(\Gamma_{\rm stim}/2)^2},$$

$$B=\frac{1}{k_i}\frac{\frac{1}{4}\Gamma_{\rm spon}\Gamma_{\rm stim}}{[E-\Delta-E(I)]^2+[\frac{1}{2}(\Gamma_{\rm spon}+\Gamma_{\rm stim})]^2}. \tag{10}$$

In this equation $\Gamma_{\rm spon}$ stands for the molecular spontaneous emission rate, taken as equal to twice the atomic rate. $\Gamma_{\rm stim}$ represents the stimulated rate for populating level e from level g, which can be estimated from the Fermi golden rule as [16,17]

$$\Gamma_{\rm stim}=2\pi\left(\frac{2\pi I}{c}\right)d_M^2|\langle f_g|n_e\rangle|^2. \tag{11}$$

This rate involves the Franck-Condon overlap between the (energy-normalized) regular wave function f_g in the ground state, evaluated at the trap energy E, and the bound-state wave function n_e in the excited state. $E(I)$ represents an energy- and intensity-dependent frequency shift, which can be estimated semianalytically [15]. We prefer, however, to consider a more operational procedure for adjusting scattering lengths, by referring to Δ as the detuning from the *observed* PA resonance, and setting $E(I)=0$ in the following. Note finally that the scattering length in Eq. (10) reduces to its ''natural'' value A_0 when the laser is switched off, setting $\Gamma_{\rm stim}=0$.

We remark that A and B are related, but not identical, to the real and imaginary parts of scattering amplitude. In particular, A emerges from the *ratio* of imaginary and real parts of the scattering amplitude (4). Accordingly, A and B do not satisfy the usual Kramers-Kronig relations for scattering in dispersive media. They do, however, satisfy an approximate dispersion relation away from resonance,

$$(A-A_0)^2+(B-B_0)^2=B_0^2, \tag{12}$$

i.e., an arc of a circle with the intensity-dependent radius $B_0\equiv(1/2k_i)\Gamma_{\rm stim}/\Gamma_{\rm spon}$. This relation will prove useful in visualizing the laser's effect, as illustrated below.

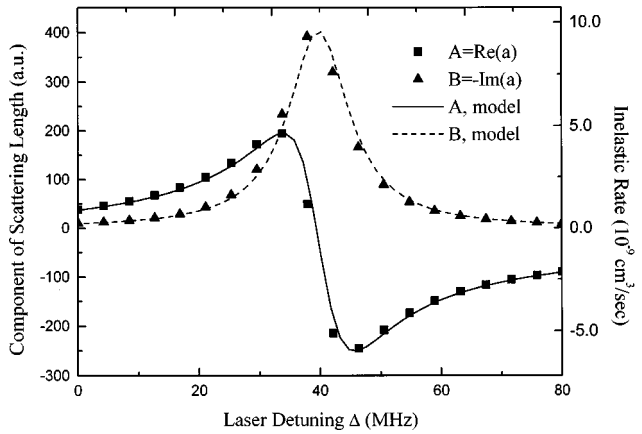

FIG. 2. Real (A) and imaginary (B) parts of the scattering length $a=A-iB$, vs detuning Δ from the $v=72$ vibrational level of the ^{7}Li excited state. The photoassociation laser has an intensity of 100 W/cm^2, and the calculation assumes a scattering energy of 1 μK. The points indicate the results of a close-coupled calculation, while the lines represent model formulas (10). The right-hand axis translates B into a two-body loss rate.

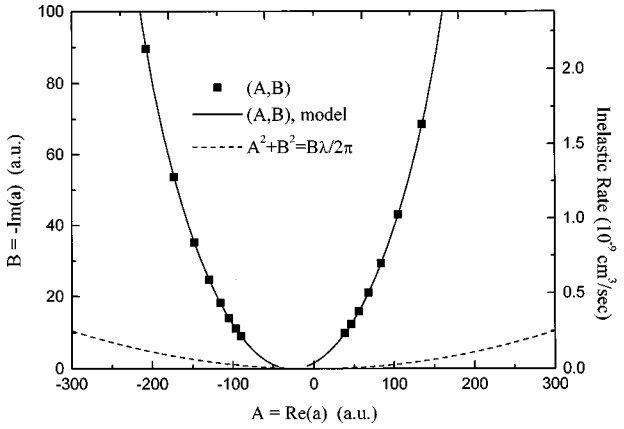

FIG. 3. We recast the results of Fig. 2 as a plot of B vs A, illustrating the approximate dispersion relation (12) as the laser's detuning varies. The points again indicate the close-coupled results, and the solid line the model. The dashed line plots the circle $A^2+B^2=B\lambda$. To ensure $K_{\rm el}>K_{\rm inel}$, the pair (A,B) must lie outside this circle.

IV. RESULTS

In this section we will present some sample results illustrating the formulas of Sec. III, for experimentally accessible circumstances. We present results for ^{7}Li and for ^{87}Rb, arbitrarily assuming a trap temperature of 1 μK.

A. ^{7}Li

As an initial application, we investigate the case of ^{7}Li, as was done in Ref. [9]. Our ^{7}Li potentials are derived from Ref. [18] for the ground-state $g={}^3\Sigma_u^+$ potential and from Ref. [19] for the excited-state $e=1{}^3\Sigma_g^+$ potential, respectively. The ground-state potential has been adjusted to have a natural scattering length of -27.3 a.u., in accordance with its measured value [20]. We choose the $v=72$ vibrational level of the excited potential as a target for the photoassociation laser, since this level yields a local maximum of the free-bound Franck-Condon factor, meaning from Eq. (11) that the applied laser has to do less work. In addition, this level lies at a detuning of $\Delta_A\sim-524$ GHz from atomic resonance, implying that the photon recoil rate (9) will remain manageable.

Figure 2 presents a plot of A and B versus laser detuning for an applied laser field of modest intensity (100 W/cm^2), and assuming a trap temperature of 1 μK. In this figure the continuous curves were derived from Eqs. (10) (including the semianalytic energy shift), while the points result from the full close-coupling calculation. This comparison demonstrates the adequacy of the model, especially away from resonance, where its results are most useful for altering scattering lengths. The imaginary component B of the scattering length shows a characteristic Lorentzian-like shape of B near resonance, while its real part A vanishes exactly on resonance.

The results in Fig. 2 resemble those of Ref. [9], which pertain to similar circumstances; as in that reference, it appears that a huge change of scattering length can be attained with a modest laser power. The photon recoil rate is under control at this intensity, accounting for the loss of only ~4 atoms/s from the trap. This is the good news. The bad news is given on the right-hand axis of Fig. 2, where the imaginary part of the scattering length is recast as a rate coefficient for inelastic scattering via Eq. (7). These enormous loss rates ($\sim10^{-9}$ cm^3/s) greatly exceed losses from any naturally occurring process in these clouds, severely limiting the utility of laser-controlled scattering lengths in this case.

The useful approximate relation (12) between A and B is readily visualized by plotting a's development in the A-B plane, as Fig. 3 does for the same conditions as in Fig. 2. For purposes of orientation, recall that this curve's minimum, where $B=0$, represents asymptotically large detunings Δ. Again the imaginary part B is scaled both as a length and as an inelastic rate coefficient. The points map the full close-coupled solution, while the solid line traces the result of approximate solutions (10). Figure 3 also shows, as a dashed line, the boundary surface $A^2+B^2=B\lambda$ implied by Eq. (8), for a trap temperature of 1 μK. For the change in A to have an appreciable effect in the atoms' mean-field interaction without destroying the cloud, the pair (A,B) must lie *outside* this circle, which is never the case in Fig. 3. Thus this example of $I=100$ W/cm^2, which looks promising at first, actually is of little use for evaporatively cooling the cloud. However, the cloud can be kicked by influencing all collisions that occur within the duration of a light pulse. Again, the specifics of the experiment must decide what is useful.

More generally, a useful rule of thumb for satisfying $K_{\rm el}\gg K_{\rm inel}$ is that the radius of the circle described by the approximate dispersion relation (12) must exceed the radius of the circle $A^2+B^2=B\lambda$, i.e., that

$$\Gamma_{\rm stim}\gg\Gamma_{\rm spon}. \tag{13}$$

This criterion asserts that the rate $\Gamma_{\rm stim}$ for driving the atom pair back into the incident channel must exceed the lossy rate $\Gamma_{\rm spon}$ for pushing the pair into inelastic channels. Since $\Gamma_{\rm stim}$ grows with laser intensity, Eq. (13) provides an estimate of the minimum intensity required to change the scat-

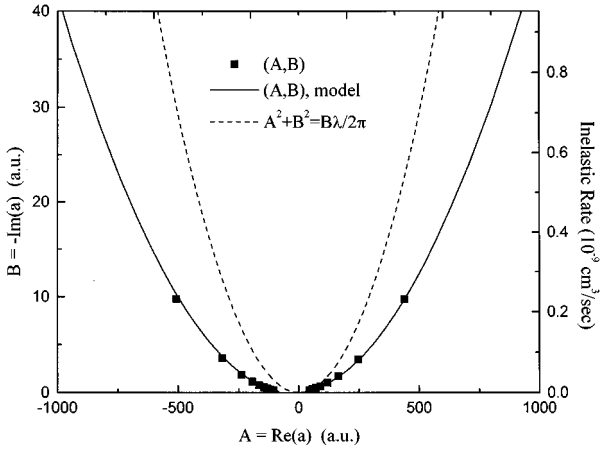

FIG. 4. Same as Fig. 3, but with the laser intensity increased to 5000 W/cm^2.

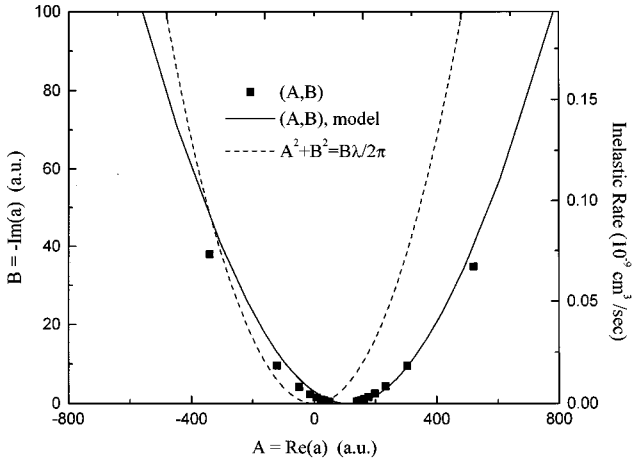

FIG. 5. For ^{87}Rb, the development in the A-B plane analogous to Figs. 3 and 4. Here the photoassociation laser is tuned near the $v=152$ vibrational level of the ^{87}Rb excited state, and has an intensity of 2000 W/cm^2.

tering length in a useful way. For the $v=72$ excited state considered here, intensities $I \geqslant 2000$ W/cm^2 meet this condition.

Accordingly, in Fig. 4 we show a similar plot in the A-B plane, with the laser's intensity boosted to 5000 W/cm^2. In this case the points (A,B) denoting the altered scattering length do lie outside the boundary $A^2+B^2=B\lambda$, indicating that elastic collisions can be driven more rapidly than inelastic collisions in this case. To take a concrete example, consider an experiment designed to create a positive sign of the ^{7}Li scattering length, making it $+50$ a.u. The model dictates that this is possible with $I=5000$ W/cm^2, leading to an imaginary part $B=0.27$ a.u., or $K_{\rm inel}=6.4\times10^{-12}$ cm^3/s, while $K_{\rm el}=7.0\times10^{-12}$ cm^3/s. To attain these conditions it is required that the laser be detuned 1.8 GHz to the red of the molecular transition, which should be possible by detuning from the lowest-energy member of the hyperfine manifold associated with the $v=72$ level. Note also that this increased laser intensity boosts the photon recoil rate 50-fold, to $\gamma_{\rm atom}\sim175$/s. Therefore, depending on the acceptable loss rate, a large change in effective scattering length can be attained with a powerful enough laser.

B. ^{87}Rb

Our ^{87}Rb $^3\Sigma_u^+$ ground-state potential was extracted from the *ab initio* potentials of Ref. [21], and our excited $1g$ potential from Amiot's [22] reconstruction of Heinzen's spectroscopic data [23]. The natural scattering length of the ground-state potential is set at 110 a.u. [24]. Here we select the $v=152$ level of the $1g$ potential as a target for the PA laser. As above, we present results for traps at a temperature of 1 μK.

At this temperature the laser intensity required to satisfy the rule of thumb (13) is ~1100 W/cm^2. We present results for a slightly higher intensity, 2000 W/cm^2, in Fig. 5. At this intensity the atomic photon recoil rate is ~60/s. Again the close-coupling calculation (squares) agrees well with model (10) (solid line). Note also the scale on the right-hand side of the graph, which shows much lower inelastic rates than for ^{7}Li. For the same temperature, ^{87}Rb will possess lower inelastic rates than ^{7}Li, owing to the mass scaling in Eq. (7). As above, the dashed line shows the circle $A^2+B^2=B\lambda$. At this laser intensity there is plenty of opportunity for the pair (A,B) to lie outside this circle, meaning that $K_{\rm el}\gg K_{\rm inel}$ for a large range of detunings.

Also, since the natural scattering length of ^{87}Rb is already as large as 110 a.u., it is much easier to satisfy Eq. (8) while *increasing* A rather than decreasing it. In the example at hand, we suppose that we want to boost A to 200 a.u. The model predicts that this will be possible at $I=2000$ W/cm^3, yielding $B=2.0$ a.u., $K_{\rm inel}=3.9\times10^{-12}$ cm^3/s, and $K_{\rm el}=3.2\times10^{-11}$ cm^3/s. This requires the laser to be detuned 0.27 GHz to the red of the molecular resonance. To make the scattering length negative presents a greater challenge, but the data in the figure illustrate that the usefulness condition (8) can be satisfied at this intensity by seeking $A<-400$ a.u.

V. SUMMARY

We have seen that a sufficiently powerful laser (in the kW/cm^2 range) can produce significant changes in scattering lengths of ultracold alkali-metal atoms. The trick is to park the laser frequency near enough to a resonance to change the scattering length without losing a host of atoms to spontaneous emission of the excited molecular state. This situation becomes possible when the rate of ''good'' (i.e., elastic) collisions exceeds the rate of ''bad'' (inelastic) collisions, as expressed by rule of thumb (13). When this condition is met, model formulas (10) can be used as a guide to how much intensity and what detuning are required to produce a desired A while limiting the size of B.

One other aspect of this problem deserves consideration, if only to mention its unfeasibility. We might consider applying a second laser to the atomic cloud, tuned on resonance between the excited molecular state and a vibrational state below, in the ground-state potential (which is stable, having no lower level in which to decay). Such two-color processes have been successful in probing the spectroscopy of the ground-state potential [20]. This circumstance produces a ''dark spot,'' a detuning for which the inelastic rate vanishes altogether [17]. Unfortunately, a semianalytic treatment of

this case, similar to Eq. (10), points out that, at the dark spot's detuning, the real part of the scattering length A returns exactly to its unperturbed value.

Actually, this is true only for the *resonant* part of the intensity-dependent a. We might expect that at sufficiently high intensity the molecular potentials themselves begin to distort, producing a concomitant distortion of scattering lengths. We performed a number of close-coupling calculations for ^{87}Rb, and verified that at kW/cm^2 intensities A can be shifted by up to 10 a.u., while B can still be made to vanish. However, to do so would require the laser to be tuned accurately to well within 1 MHz, making this approach seem hardly worthwhile.

ACKNOWLEDGMENTS

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