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Novel Intensity Dependence of Ultracold Collisions Involving Repulsive States.

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Abstract. – We present measurements of the intensity dependence of excited-state collisions of laser-cooled ^{85}Rb atoms involving repulsive excited-state potentials. For high intensities, the collision rate decreases with increasing intensity, in accordance with a simple Landau-Zener treatment of the collision dynamics.

Interest in excited-state collisions of laser-cooled atoms stems from the extreme sensitivity of the atoms to weak long-range interatomic forces and the similarity of collisional and radiative time scales [1]. In particular, since the interatomic forces differ by orders of magnitude when the atoms are in the ground or excited electronic states, the collision dynamics are strongly affected by interaction with radiation, with both stimulated and spontaneous processes being important. Proper understanding of the collision dynamics, including the effects of spontaneous processes, is a challenging problem that is central to ultracold-collision phenomena.

In a first attempt to understand ultracold-collision dynamics, Gallagher and Pritchard [2] proposed that the collisions could be understood as a sequential process. A colliding pair of atoms in their ground states absorb a photon to an attractive excited-state potential. Acceleration of the atoms toward each other by the excited-state potential then results in inelastic energy transfer, unless spontaneous emission occurs before the atoms come sufficiently close to each other for the energy transfer to occur. The collision rates are therefore taken to be a product of three factors: the excitation rates (calculated using a quasi-static theory), survival probabilities, and energy transfer probabilities. Meaningful experimental tests of the predictions that follow from these ideas have been frustrated by the complications introduced by hyperfine interactions, although progress along these lines has recently been reported [3].

Recently, fully quantum-mechanical model calculations of the collision dynamics have been made that suggest revisions to this picture [4]. These calculations are well represented by a model in which separability of the excitation and survival probabilities is retained, but the excitation probability is found to be well described by a simple Landau-Zener expression, similar to that explained below. It is crucial to experimentally test these ideas. As a first step, in this paper we present measurements of the laser intensity dependence of ultracold-collision rate involving *repulsive* molecular states. These collisions are of particular interest, because for this situation the Landau-Zener model predicts that at high intensities the collision rates *decrease* with increasing intensity. Indeed we observe this effect, and find it in quantitative agreement with the model.

The trap-loss mechanism for repulsive states is depicted in fig. 1. Two colliding atoms are excited by a laser field of frequency ν_L to the high-frequency side of the atomic resonance frequency ν_0. If $\Delta = \nu_L - \nu_0$ is large enough (typically 10 GHz or more), the atoms are ejected from the trap after being rapidly accelerated by the steep excited-state potential. For these detunings the atoms are accelerated sufficiently rapidly that spontaneous emission effects can be neglected.

A Landau-Zener model of this process treats the excitation probability as a curve crossing between two diabatic states of the atom-pair–laser-field system, $|g; N\rangle$ and $|e; N-1\rangle$, where «g» and «e» denote ground and excited states of the atom pair, and N the number of photons in the laser field. The interaction potentials for the two states are $V_g = h\Delta$ and $V_e = C_3/R^3$, where R is the interatomic separation (for the moment we ignore the presence of multiple potential curves). Coupling between the two states is given by $h\varepsilon$, where ε is the Rabi frequency. The collision begins with incoming atoms in the state $|g; N\rangle$. A curve crossing occurs at interatomic separation $R = (C_3/h\Delta)^{1/3}$. The Landau-Zener treatment then gives the transition probability to the excited-state curve as

$$P = 1 - \exp[-\gamma(b, v_r)], \qquad \gamma(b, v_r) = 8\pi^3 \varepsilon^2 C_3^{1/3}/3v_r(h\Delta)^{4/3}, \tag{1}$$

where b is the impact parameter for the collision, and $v_r = v\sqrt{1 - b^2/R^2}$ is the radial velocity at the crossing point. Once excited, the radial velocity is quickly reversed by the excited-state potential and the atoms pass through the curve crossing again, remaining in the excited state with a probability $1 - P$. Subsequent acceleration by the excited-state potential gives the atoms sufficient kinetic energy to escape the trap. The trap-loss probability is therefore $P(1 - P)$. Alternatively, if the incoming atoms are not excited at the crossing point (probability $1 - P$), they reflect off the centrifugal barrier of the ground-state potential and return to the crossing point where they are excited with probability P. Thus the total trap-loss probability is the usual Landau-Zener result $P_t = 2P(1 - P)$.

Since $\gamma \propto \varepsilon^2$ and hence is proportional to laser intensity, this predicts that for large intensities the trap-loss rate will *decrease* with increasing intensities, instead of saturating as

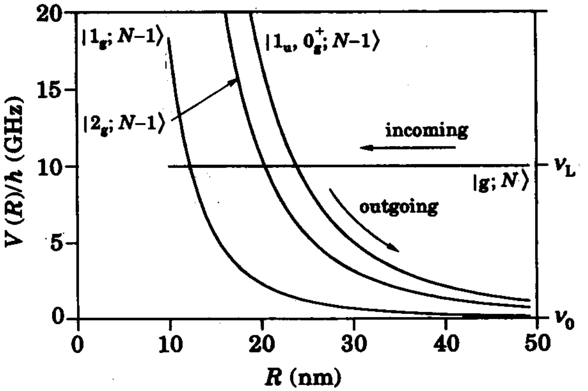

Fig. 1. – Potential curves for repulsive trap-loss collisions.

most optical processes do. This is a striking prediciton of the Landau-Zener model, and is quantitatively confirmed by the measurements we now describe.

The principles behind the technique used to measure the collision rates have been described elsewhere [5], with minor variations. A ^{85}Rb magneto-optical trap was used with a trap laser intensity of 2 mW/cm^2, detuning of -4.9 MHz from the $5S_{1/2}(F=3)\to 5P_{3/2}(F=4)$ transition (frequency ν_0), and beam waist of 6.3 mm. The magnetic-field gradient was 20 G/cm. Under these conditions, the MOT is known [6] to have a trap depth of > 7 GHz. In fact, we have used the repulsive trap-loss collisions to measure that the trap depth is ≈ 10 GHz (with considerable anisotropy), as will be described in a forthcoming publication [7]. The trap was loaded directly from a background Rb vapor, and the number of trapped atoms, as deduced from fluorescence measurements, was typically $3\cdot 10^6$, with typical densities of $3\cdot 10^{10}$ cm^{-3}. The repulsive-state collisions were induced by adding an intense, «catalysis» laser to the trap whose detuning Δ was varied from 5–20 GHz to the blue of ν_0. For reasons described below, this laser (intensity 1–15 W/cm^2) was chopped at a frequency of 1 kHz with a variable duty cycle d. In order to avoid standing waves, the catalysis laser beam was not retroreflected. The collisions induced by the catalysis laser reduce the number of trapped atoms N, since N depends on a balance between the loading rate and the collisional loss rate: $N = L/(\Gamma_t + \langle\beta n\rangle d)$, where L is the loading rate, Γ_t the loss rate due to collisions other than those induced by the catalysis laser, and $\langle\beta n\rangle$ is the spatially averaged collision rate induced by the catalysis laser.

Since $\langle\beta n\rangle$ depends on the density distribution in the trap, which in turn depends on N, we preferred to adjust the duty cycle d in order to hold N constant as the catalysis laser intensity varied. Then β is inversely proportional to the duty cycle required to hold N constant. This eliminated the necessity of making detailed measurements of the density distribution. However, AC Stark shift induced by the catalysis laser must be taken into consideration. Because of the AC Stark shift, the scattering rate from the trapping lasers changed when the catalysis laser was on, so we deduced N from the fluorescence only during the time periods when it was off. It was also necessary to chop the catalysis laser beam at a frequency of 500 Hz or greater to minimize AC-Stark–induced changes in the shape of the density distribution. We also checked for a change in the loading rate produced by the catalysis laser, but experimentally we found no such effect at the 5% level.

Our measurements of the intensity dependence of β at a catalysis laser detuning of 10 GHz are shown in fig. 2*a*). As predicted by the Landau-Zener model, the trap-loss rates as a function of intensity show a clear peak and decrease at large intensities.

A detailed comparison of the experimental results to theoretical predictions is beyond the

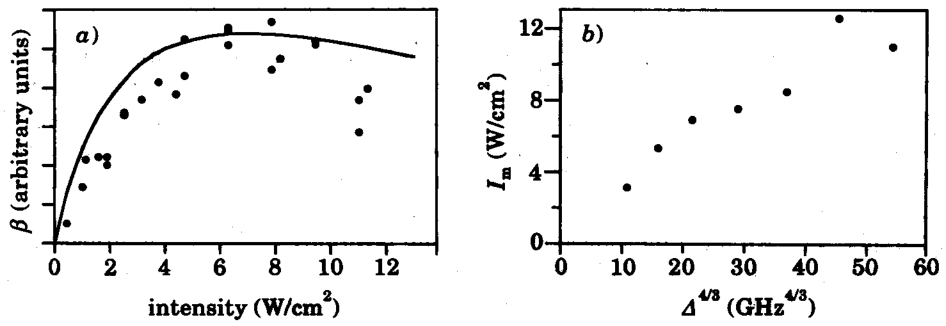

Fig. 2. – Measurements of collision rates for repulsive trap-loss collisions. *a*) Collision rate as a function of catalysis laser intensity. *b*) The position of the maximum of curves such as *a*) as a function of detuning.

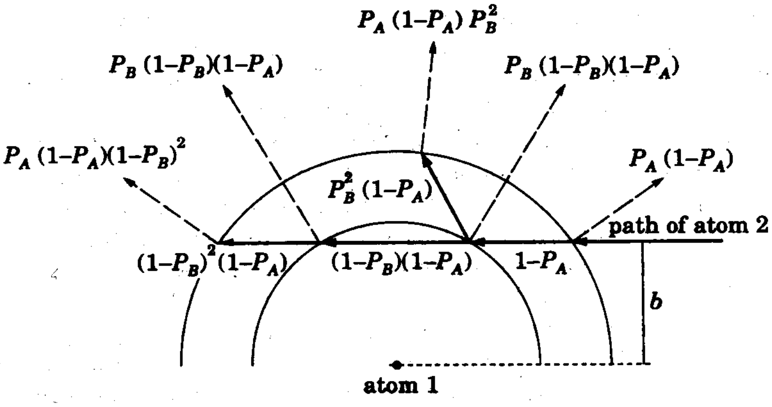

Fig. 3. – Trap-loss pathways for the case of two excited-state potentials, denoted by «A» and «B». The partial circles represent interatomic separations where the laser is in resonance with the colliding atoms. Motion on ground-state curves is denoted by dark arrows, motion on excited-state curves by dashed arrows. The relevant probabilities for each pathway are also shown.

scope of this paper. However, we have made estimates based on the following simplified model. First, including fine structure but neglecting hyperfine structure, there are only four molecular curves [0_g^+(oscillator strength $f = 0.083$), $1_u(0.198)$, $2_g(0.5)$, and $1_g(0.365)$] that are repulsive and have radiatively allowed dipole transitions to the ground state [8]. At the detunings used in this experiment the 0_g^+ and 1_u states are strongly mixed by the hyperfine interaction, so for simplicity we consider them as a single state with total oscillator strength equal to the sum of the individual oscillator strengths. We then have a model with three excited-state potential curves shown in fig. 1.

There are many new collision pathways that result in trap-loss for the case of several excited-state potential curves, as illustrated in fig. 3 for the case of two potential curves. For example, if the atom pair is not excited at the crossing with the potential curve A (probability $1 - P_A$, where P_A is determined analogously to eq. (1), taking into account the oscillator strength of the transition), it can be excited at the second crossing (probability P_B), be turned around by the repulsive potential, then immediately be de-excited on the second traversal of curve crossing B (probability P_B), and then finally be excited on the second traversal of curve A (probability P_A). Thus, the trap-loss probability for this pathway is $P_A(1-P_A)P_B^2$. Summing all such pathways, we get a total trap-loss probability

$$P_t = \{P_A[P_B^2 + (1-P_B)^2] + 2P_B(1-P_B)\}(1-P_A). \tag{2}$$

The $(1 - P_A)$ factor in eq. (2) is primarily responsible for the decreasing intensity dependence at high intensities. To obtain the total trap-loss rate, we integrate the analogous formula for three potential curves over impact parameters and average over the velocity distribution. The result, with only the overall vertical scale factor being treated as an adjustable parameter, is the solid line in fig. 2. The data and the model agree on the position of the maximum, but the peak is sharper in the data than in the model calculation.

As a further test, we have measured (fig. 2*b*)) the positions $I_m(\Delta)$ of the intensity maxima of curves like fig. 2 for various detunings. From eq. (1), the detuning dependence should obey $I_m \propto \Delta^{4/3}$, and the data are consistent with this prediction.

We have shown that the Landau-Zener prediction for the intensity dependence of trap-loss collisions involving repulsive states accounts for our measurements fairly well. In order to test the models [4] better, it would be desirable to study collisions with smaller detunings, where spontaneous-emission effects would be more important. This might be done, for example, using a weaker trap, or by studying collisional suppression of ground-state hyperfine-changing collisions [9].

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