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Direct measurement of fine-structure collisional losses from a Cs magneto-optical trap

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Losses from a cesium magneto-optical trap induced by fine-structure changing collisions have been measured directly by counting of fluorescence photons emitted at the D_1 resonance line, with the trap lasers operating on the D_2 resonance line. The absolute value of the fine-structure loss rate coefficient has been measured and its dependence on the hyperfine state occupation has been explored. These results provide a fundamental test for the theoretical descriptions of excited-state trap losses. [S1050-2947(97)50906-1]

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Cold atomic samples produced by laser and evaporative cooling provide an opportunity for a wide range of experiments, including those on Bose-Einstein condensation. A great deal of interest in cold atoms has also been stimulated by the unusual features accompanying collisions between cold atoms [1,2]. In an inelastic collision an energy transfer between the two colliding atoms may take place, and one, or both atoms, may gain enough kinetic energy to escape the potential barrier defining the trap. In so-called optical collisions one of the colliding atoms is excited to an attractive potential curve. Optical collisions constitute the main loss mechanism from an optical trap, where the confining potential originates from the laser field. For alkali-metal atoms fine-structure (FS) changing collisions and radiative escape (RE) collisions are the dominant optical collisions. In FS collisions an atom enters the collision in the $P_{3/2}$ excited state and exits in the $P_{1/2}$ state, the energy difference being converted to kinetic energy of the colliding atoms. The interaction between colliding atoms is determined by the molecular potential, precise knowledge of which, including the hyperfine structure, is required for the interpretation of the processes [1,3-5]. FS optical collisions can be viewed at weak laser fields as three-step molecular processes. In the first step optical excitation leads the colliding pair to an excited molecular state, asymptotically correlated to the $S_{1/2} + P_{3/2}$ limit. In the second step the excited-state population evolves along that molecular potential from the internuclear distance of excitation to the small internuclear separation at which a curve crossing between two molecular states occurs. In the final step the colliding atoms separate, again on a molecular potential curve; this time, however, on a curve that is connected to the $S_{1/2} + P_{1/2}$ asymptote.

In the present work, detection of D_1 fluorescence emitted in the $6P_{1/2} \rightarrow 6S_{1/2}$ transition by cesium atoms confined in a magneto-optical trap (MOT) operating on the D_2 , $6P_{3/2} \rightarrow 6S_{1/2}$ transition, provides direct evidence of the FS loss process. Measurement of the photon yield, together with that of the number of atoms present in the trap, allows us to derive an absolute value for the FS trap loss rate. If, as theory predicts [1,4], FS losses take place through crossings between molecular potential curves asymptotically connected to definite hyperfine atomic levels, only certain input channels should contribute. In our experiment we provide an original test of the sensitivity of FS trap losses to the input hyperfine channel by modifying the initial occupation of the hyperfine ground states, and, for a given ground-state hyperfine occupation, by populating different hyperfine excited levels. To this end, we have measured the production of D_1 photons in two different MOT laser configurations, i.e., lasers driving the $4_g \rightarrow 5_e$ and $3_g \rightarrow 4_e$ transitions (type-I MOT [6]) and lasers driving the $4_g \rightarrow 5_e$ and $3_g \rightarrow 2_e$ ones (combined type-I-II MOT [6]).

Our σ^+, σ^- MOT [7] operated with field gradient dB/dz at 0.2 T/m, and trap laser detuning $\delta_T = -1 - 5\Gamma$, where Γ is the excited-state spontaneous decay rate. High Rabi frequencies were used for both the trapping and repumping lasers, with total intensity up to $I_T = 150 \text{ mW/cm}^2$ for the trapping $4_e \rightarrow 5_e$ transition. For the type-I trap the repumping laser was at -3.6Γ from the $3_g \rightarrow 4_e$ transition and its total laser intensity ranged up to $I_R = 60 \text{ mW/cm}^2$. The type-I-II combination was produced by tuning the repumping laser at 4Γ on the red side of the $3_g \rightarrow 2_e$ transition. Particular care was taken to have both traps centered at the same spatial point. The number N of trapped atoms, ranging between 0.3×10^8 and 4.8×10^8 , was derived from the MOT fluorescence flux $I_{\rm fl}$ measured through a calibrated photodiode. The spatial distribution function, g(r,N) with g(0,N) = 1, was derived from charge-coupled-device images of the MOT. The maximum atomic density n_{max} was around $1\!-\!4\!\times\!10^{11}$ atoms/cm $^3.$ The MOT temperature was measured between 50 and 300 μ K through a time of flight. The ground hyperfine-state fractions $f_{3_{g}}$ and $f_{4_{g}}$ were derived from absorption of a probe beam. Independent and consistent values for the atomic fractions were derived for the type-I-II trap from measurements of the fluorescence decay times after switching off the trapping or repumping lasers. The background pressure p_{h} was measured from absorption of a probe laser propagating through the cesium vapor outside the MOT.

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FIG. 1. Ratio R_{D_1,D_2} of the counting rates for the type-I trap as a function of the cesium background pressure p_b , for $I_T = 150$ mW cm⁻², $\delta_T = -3\Gamma$, and repumping laser at $I_R = 60$ mW cm⁻² resonant with $3_g \rightarrow 4_e$ transition. The ratio R_{D_1,D_2} is constant for a factor of 200 increase in p_b .

A combination of a monochromator, with a red enhanced photomultiplier, and an interference filter was used to detect D_1 photons. The ratio η_{D_2/D_1} between the total detection efficiencies at the D_2 and D_1 wavelengths was measured to 7.0±0.5. The solid angle for collection was $\approx 1.3 \times 10^{-2}$ sr, and the D_2 efficiency was $\simeq 2 \times 10^{-5}$. The maximum C_{D_1} counting rate was 10 s⁻¹, with a 1-2 s⁻¹ background due to scattering of D_2 photons. The D_2 counting rate C_{D_2} was in the 10^6 counts s⁻¹ range, when measured with an optical density 3.1 absorption filter inserted at the monochromator entrance. This counting rate is in good agreement with that expected for our number of trapped atoms. Counting rates have been corrected for the absorption by background cesium atoms. Even if the C_{D_2} and C_{D_1} rates depend on the optical alignment of the detection apparatus, which could change in separate runs, their ratio $R_{D_1,D_2} = C_{D_1}/C_{D_2}$ is independent of the alignment.

Several tests were performed to ensure production of D_1 photons by cold collisions. Comparison with D_1 emission from a cesium spectral lamp verified that the apparent frequency distribution of the D_1 detected photons was determined by the spectral resolution of the monochromator. D_1 photons were present only with cold atoms present, and their production increased with the cold-atom number. D_1 photons did not originate from collisions between a cesium atom in the MOT and a cesium atom in the background, even at the largest p_b of the experiment. Figure 1 shows that increasing p_b two hundred times, R_{D_1,D_2} remains nearly constant. Furthermore, using the measured cesium background density and the cross section for FS collisions at room temperature [8], background–cold-atom collisions lead to a D_1 photon counting rate at least two orders of magnitude lower than that measured. Thus, our D_1 counting rate is certainly produced by collisions between cold cesium atoms in the MOT. The D_1 photons were not produced by collisions between excited atoms because the counting rate did not follow the laser dependence required for those collisions.

The rate equation that governs the number N of trapped atoms is [3]

$$\frac{dN}{dt} = L - \gamma N - \beta n_{max} f(N) N, \qquad (1)$$



FIG. 2. Measured β^{FS} versus *N* for a type-I trap varying p_b and the laser detuning at laser intensities as in Fig. 1. The line represents a fit for a linear dependence with intercept 1.3×10^{-11} cm³ s⁻¹ and slope 2.1×10^{-20} cm³ s⁻¹.

where *L* is the loading rate from the background vapor, and γN is the trap loss rate for collisions with the background vapor. The last term, describing the loss due to collisions between trapped atoms, is characterized by the rate coefficient β . The factor $f(N) = \int g^2(r,N) d^3r / \int g(r,N) d^3r$ measures the reduction in collision rate resulting from the spatial atomic distribution. If the optical potential barrier is large enough to inhibit the hyperfine changing losses, the loss coefficient β has contributions β^{RE} and β^{FS} from RE and FS collisions. Using $I_{\text{fl}} = f_e N \Gamma \eta_{\text{ph}}$, with f_e the excited-state fraction and η_{ph} the calibrated photodiode collection efficiency, and a similar equation for the C_{D_1}/C_{D_2} fluorescence count, the β^{FS} coefficient is expressed as

$$\beta^{\rm FS} = 2R_{D_1, D_2} \frac{\eta_{\rm ph}}{\eta_{D_1/D_2}} \frac{\Gamma^2 f_e^2}{f(N) I_{\rm ff}} \int g(r, N) d^3 r.$$
(2)

The factor 2 arises because, for each emitted D_1 photon, two atoms are lost from the trap. f_e is derived from the applied Rabi frequencies and laser detunings using a corrected saturation relation derived in [9].

 β^{FS} was derived as a function of the trap parameters from the measured values of R_{D_1, D_2} . No dependence on the trap laser detuning was found in the explored range, as expected for the strong saturation of the laser excitation. Figure 2 shows β^{FS} vs N for the type-I trap, at fixed laser intensities, by varying the background pressure p_b . For $N \simeq 4 \times 10^8$ atoms we measured the scatter in the β^{FS} values originated by different optical alignments. That scatter evidences our limit in the absolute determination of the FS loss coefficient. The straight line in Fig. 2 represents the N dependence on β^{FS} for data taken at a fixed optical alignment. The weak dependence of β^{FS} on N provides additional evidence that the FS loss rate is a cold-atom process described by Eq. (1). The remaining influence of N on β^{FS} given by the fit was ascribed to multiple scattering effects not completely described by the f(N) correction in Eq. (1). No evidence of dependence of β^{FS} on the trap temperature was observed.

Figure 3 shows experimental results for the β_{FS} dependence on the f_{3_g} fraction and Fig. 4 the dependence on the trapping laser intensity. In the type-I trap the 3_g occupation was controlled by varying the repumping laser intensity. For

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FIG. 3. β^{FS} versus f_{3_g} for a type-I (squares) and for a type-I-II trap (circles). The lines correspond to the parameters $a_T = 15$, $a_R = 1$, $\xi_0 = 0.24$, for the lower data $\beta_{0R}^{\text{FS}} = 2.7 \times 10^{-10}$ cm⁵ s⁻¹ mW⁻¹, and for the upper data $\beta_{0R}^{\text{FS}} = 0.7 \times 10^{-10}$ cm⁵ s⁻¹ mW⁻¹.

a weak repumper the MOT contained a smaller number N of trapped atoms, but a larger f_{3_g} fraction. Operation of the type-I-II trap produced an increase in the R_{D_1,D_2} ratio and therefore in the β^{FS} value, as shown by the upper data in Fig. 3. In comparing the β^{FS} values in the two traps, notice that in the type-I trap a large f_{3_g} occupation was obtained at very weak repumping laser intensities, while in the type-II trap it was obtained at repumping laser intensities comparable to the trapping ones.

A precise theoretical analysis of our data requires knowledge of the cesium molecular potentials, including hyperfine structure, at present not available. Moreover, our results apply to a regime of large trapping and repumping laser intensities, where the molecular potential crossings may be modified by the strong laser couplings. We have analyzed the β^{FS} data on the basis of a simple model that takes into account the main features of optical collision losses. We only consider long-range excitation processes, since a simple Landau-Zener model shows that direct excitation at short range of repulsive states leading to direct production of the D_1 line predicts orders of magnitude less signal than is observed. Theoretical considerations of the molecular hyperfine structure indicate that no molecular potentials that adiabatically connect with the upper set of asymptotic states $4_g + P_{3/2}$ can contribute to the FS trap-loss rate [10]. Thus, we assume that the molecular hyperfine states relevant for the FS losses connect with $3_g + 4_e$ for the type-I trap and $3_g + 2_e$ for the type-II trap, both excited by the repumping



FIG. 4. β^{FS} versus I_T for a type-I trap, at $\delta_T = -3\Gamma$, $I_R = 62$ mW cm⁻². The line corresponds to the parameters of Fig. 3.

laser from the 3_g states so that β_{FS} depend on $I_R f_{3_g}^2$. Excitation of the 3_g atoms by the trapping laser at small internuclear separation results in a $I_T f_{3_g}^2$ dependence. β^{FS} depends on the excited-state survival in the evolution along the excited molecular potential from the region of excitation to the small internuclear distance where the curve crossing that leads to the FS transition takes place. Since the exciting laser fields are quite strong, they can cause excitation in a broad region about the Condon point with the ground state, and the atomic wave packet that describes the approaching atoms undergoes several excitation and/or decay cycles. As a consequence, normal exponential decay is inhibited until the wave packet has moved far away from the excitation point [11]. Under these conditions we neglect the dependence of the survival factor on the laser intensities.

The data of Fig. 3 cannot be fitted by assuming that $f_{3_g}^2$ represents the total functional dependence of β^{FS} on the ground-state fractions. On the contrary, a good fit is obtained assuming that the population of the 4_g ground state also participates in the FS loss process through the following functional dependence:

$$\beta^{\rm FS} = \beta_{0R}^{\rm FS} \frac{I_R}{1 + I_R / (a_R I^s)} [f_{3_g}^2 + \xi f_{3_g} f_{4_g}]$$
(3)

with

$$\xi = \xi_0 \frac{I_T / (a_T I^s)}{1 + I_T / (a_T I^s)}.$$
(4)

The β_{0R}^{FS} term describes the f_{3_g} excitation by the repumping laser at large internuclear distances. The coefficient ξ represents the probability that within a single collision, near the initial Condon point, an optical pumping process transfers the 4_g occupation to the 3_g state. Thus, owing to an optical pumping process, the hyperfine occupations of the atoms contributing to the FS losses are different from the average ones. An additional $\xi^2 f_{4_n}^2$ contribution, describing the FS loss contribution of two atoms initially in the 4_g state and optically pumped into the 3_g state, is negligible for our range of parameters. Equation (3) assumes that these optically pumped atoms contribute to the FS losses via the same channels as atoms originally in the 3_g state. Different loss channels or new channels associated with the strong laser modifications of the molecular potential crossings may be also assumed in the functional dependence of β^{FS} , but for the present data the contributions of those mechanisms could not be resolved because they are too small. The a_i^s parameters, with (i=R,T), represent the molecular correction to the atomic saturation intensity I_s , the molecular excitation rate not saturating as readily as the free-atom excitation [12]. Equation (3), with five free parameters, was used to fit simultaneously the data of Figs. 3 and 4, corrected for the dependence of β^{FS} on N shown in Fig. 2. We have fitted the loss data of the two traps by changing only the β_{0R}^{FS} parameter describing the excitation by the repumping laser, whose frequency was different. An $I_T f_{3_a}^2$ term describing the excitation by the trapping laser at small internuclear distances, if introduced into the fit, results in a contribution that is negliR4002

gible compared to the contribution from large distances. Therefore the FS losses are produced by the repumping laser excitation at large internuclear distances.

The main results of our analyses of the FS losses are the estimate of the FS losses and their dependence on the trap parameters. At trap laser total intensity of 10 mW cm⁻², we obtain β^{FS} about 2×10^{-12} in our type-I trap. From a comparison with the total loss rate coefficient β measured in Refs. [13-15], we deduce that the FS loss causes one-fourth of the total losses, i.e., the RE rate is larger than the FS rate by roughly a factor of 3. Reference [1], which neglected hyperfine structure, estimated that for cesium the FS rate should be twice the RE rate. Our results show that the hyperfine structure plays a prominent role. Although the parameters accessible in the experiment may modify simultaneously the trap operation and the FS losses, and these two effects are difficult to disentangle, our model provides several important indications on the dependencies of the FS losses. These losses are produced by collisions between atoms in the 3_{g} ground state excited by the repumping laser. Excitation by the trap laser appears to be not important, but optical pumping by the trap laser opens the possibility for atoms in 4g state to contribute to FS losses. Application of our results to an analysis of other experiments requires a precise knowledge of the trap parameters of each experiment, because β^{FS} depends on both hyperfine fractions and

on trapping and repumping laser intensities. For instance, the large 3_g occupation realized in the dark-spot traps implies that FS losses may play an important role in those traps. However, that role could be controlled by a proper choice of the trapping and repumping laser intensities.

In summary, we have reported an observation of FS changing cold collisions, which in previous experiments were indirectly deduced by measuring the total trap losses. A careful analysis of the atom spatial distribution systematically applied, for each operation of the trap, has allowed us to include the main effects of multiple-scattering through the f(N) term. The remaining dependence of the loss rate on the number of trapped atoms requires further theoretical analysis of the multiple scattering process. Our direct measurement of β^{FS} provides a challenge to improve the theoretical calculations for the cold-collision processes. Any progress in the identification of the detailed cesium loss mechanisms may permit a reduction of the trap losses through optical suppression of the collisions.

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