Population Dynamics in Coherent Excitation of Cold Atoms

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Introduction

A summary of the entire scope of the Kansas State University AMO program is contained in the J. R. Macdonald Laboratory abstract. The present document is limited to a discussion of recent progress in the MOTRIMS projects.

MOTRIMS, or magneto optical trap recoil ion momentum spectroscopy, is a methodology originally developed for the study of ion-atom or photon-atom collisions. Following the basic “inverse kinematics” idea underlying COLTRIMS (cold target recoil ion momentum spectroscopy) MOTRIMS combines well-established techniques for cooling and trapping atoms with the equally well-established techniques for measuring the momenta of product ions in the above-mentioned collision types. Several years ago it was realized that the whole idea of MOTRIMS could be turned around and charge transfer is now being used as a probe of population dynamics in cold, trapped atomic samples.

The key idea behind MOTRIMS is that in an ionizing collision, the recoiling target ion’s momentum carries with it detailed information about the collision process. For example, in the single electron transfer process of this work, the Q-value, or energy defect of a collision is given by

\[ Q = -\frac{1}{2} m_e v_p^2 - p_{\parallel} v_p, \]  

(1)

where \( m_e \) is the mass of the electron, \( v_p \) is the velocity of the projectile, and \( p_{\parallel} \) is the component of the recoil ion velocity that lies parallel to the initial direction of the projectile ion. The energy defect, \( Q \), can be thought of as the difference between the initial and final potential energies of the transferred electron. (The transverse component of the recoil momentum is related to the projectile scattering angle, but that is not relevant to the studies described here.) So, a measurement of the recoil ion’s \( p_{\parallel} \) gives the Q-value of the collision which, in turn, gives the initial and final states associated with that collision. Taken together with the previously measured relative cross sections of the different collision channels, a measurement of the relative count rates for the different collision channels then yields the relative initial populations of the different states of the target before the collision. For example, for a 2-level system in which the two states of the system are designated by the subscripts \( s \) and \( p \), the fraction of atoms in the \( p \) state is given by

\[ f_p = \frac{n_p}{n_s + n_p} = \frac{A_p/\sigma_p}{A_s/\sigma_s + A_p/\sigma_p} = \frac{A_p}{RA_s + A_p}, \]  

(2)
where \( n_s \) and \( n_p \) are the relative numbers of atoms in the indicated states, \( A_s \) and \( A_p \) are the measured rates for charge transfer from the indicated initial states, and \( \sigma_s \) and \( \sigma_p \) are the relative cross sections for charge transfer from the indicated states. The term \( R \) is defined as \( \sigma_p/\sigma_s \).

In the remaining sections of this abstract, a brief summary of two MOTRIMS projects will be given. In the first, the steady-state, average excited fraction in a MOT was measured as a function of trapping laser detuning and intensity. The results were compared with predictions of simple models. In the second project, the population dynamics of a 3-level ladder system undergoing coherent 2-color excitation was measured.

**Excited State Fraction in an “Active” Magneto Optical Trap**

At some time or other most groups using a MOT need to know the what fraction of atoms in the MOT are in an excited state. This is because the most convenient way to determine the total number of atoms in a MOT is to make a measurement of the total number of atoms in the excited state (via a fluorescence measurement) and then to divide this number by the excited fraction. Knowing the total number of atoms in a MOT is important for several reasons. The most common one is so that one can determine the density of atoms in the MOT. Researchers are interested in knowing this because many processes, including cold collisions, depend on the density. Until now, however, the MOT community has had to rely on untested simple models to estimate the excited fraction of atoms in their MOTs. The most commonly used expression for the excited fraction is

\[
f_{ex} = \frac{I/I_s}{1 + 2I/I_s + (2\delta/\Gamma)^2},
\]

where \( I \) is the total trapping laser intensity (the incoherent sum of the intensities of all the trapping laser beams), \( \delta \) is the trapping laser’s detuning from resonance, \( \Gamma \) is the full linewidth of the atomic transition, and \( I_s \) is the so-called saturation intensity, often given by

\[
I_s = \frac{2\pi h \epsilon \Gamma}{3\lambda^3},
\]

where \( h \) is Planck’s constant and \( \lambda \) is the transition wavelength. Note that Eq. (3) does not contain dependencies on several important MOT parameters, including B-field gradient, repump laser intensity, and the intensity balance between the different trapping laser beams. This equation is exact for a single travelling optical wave, interacting with a true 2-level system, in the absence of external fields, and at low enough target densities so as to be able to neglect radiation trapping. However, in a MOT one typically does not have a 2-level system due to the presence of Zeemann splitting from the magnetic field gradient. Furthermore, radiation trapping is nearly always taking place, and instead of a single travelling wave, the atoms in a MOT are typically subjected to 3 pairs of counter-propagating waves whose relative phases are typically not fixed. Thus, in a MOT one typically has spatially and temporally varying pump laser beam intensities interacting with a spatially-varying Zeemann-split system, of unknown m-distribution that can trap some fraction of the spontaneously emitted radiation. Under these conditions, the use of Eq. (3) as an accurate predictor of excited fraction does not seem promising. Nevertheless, an experiment was carried out using the MOTRIMS apparatus to study \(^{87}\text{Rb}\), in which the excited state fraction was directly measured, while the trap laser detuning and total intensity were continuously varied. Remarkably, it was found that Eq. (3) does indeed work, over a wide range of B-field gradients, intensity balances of the trapping laser beams, and repump laser intensities. However, instead of using Eq. (4), which gives a value of 3.2 mW/cm\(^2\) for \( I_s \), a fitted value of 9.2 mW/cm\(^2\) had to be used. Using this value...
for $I_s$ in Eq. (3), along with the measured trap laser detuning and total intensity, our study shows that one can confidently estimate the excited fraction of $^{87}$Rb in a MOT to within $\pm 1\%$.

**Population Dynamics During STIRAP**

Developing the ability to efficiently control populations would benefit many areas of science and technology. For example, if one could efficiently place reactants in certain states, one could potentially control the speed of a desired chemical reaction. In the area of quantum information, much depends on the efficiency with which one can create or interact with individual qubits. Over the past decade, one of the developments in coherent interaction of light with matter is the particular excitation scheme known as stimulated Raman adiabatic passage, or STIRAP. Although both theory and experiment have made progress in characterizing STIRAP, until now the temporal evolution of populations in a system undergoing this form of excitation has not been observed. Now, using the methodology described above, such population dynamics have been observed on the few nanosecond time scale.

In the experimental procedure, $^{87}$Rb was trapped and cooled in the usual manner. The trapping lasers were then turned off for a period of 500 ns, allowing the excited atoms to decay to the ground state. Then, while the trapping lasers were still off, 50 ns pulses of optical excitation were allowed to interact with the rubidium, one tuned just to the red of the Rb($5s$) → Rb($5p$) transition, and the other tuned to the blue, by the same amount, of the Rb($5p$) → Rb($4d$) transition. This process was repeated at a frequency of 200 kHz with the continuous accumulation of Q-value spectra. The resulting 2-dimensional spectra (counts versus Q-value and time) were then analyzed by dividing the number of counts in each charge capture channel by the corresponding relative cross section, as described above in the section on MOT excitation fraction. In this case, there were 3 levels of interest: Rb($5s$), Rb($5p$), and Rb($4d$). The relative excited state fractions were normalized to unity and plotted versus time.

In this excitation scheme there were essentially 5 experimental parameters that could be varied. These are the peak intensities of the two optical excitation pulses, the temporal widths of the pulses, and the relative timing between these pulses. In the work presented here, only the intensities and relative timing were varied. Because the temporal evolution of all three levels were measured, the effects of changing these parameters were readily observed, including the effects of changing the excitation from the completely adiabatic to partially diabatic regimes.

**Summary**

The emphasis of the work presented here has been on measuring the relative populations of atoms that have been cooled and trapped in a MOT. In one case, the temporal evolution of the populations were measured with a resolution of a few nanoseconds. Although it is believed that the examples presented here are already of great value to the greater scientific community, it is also believed that this represents only the beginning of what will eventually be accomplished using the MOTRIMS methodology. For example, presently in the Macdonald lab experiments are under way in which photo-association with subsequent excitation along a ladder of molecular states is being measured. In addition, a second MOTRIMS apparatus has been constructed and installed on the Macdonald Lab EBIS, under the direction of Dr. C. Fehrenbach. This will allow more flexibility in the choice of projectile ions used in this sort of measurement. For example, until now, only alkali ions have been used with MOTRIMS to probe population dynamics. However, due to spin-orbit interactions lifting the $l$-degeneracy in the projectile energy structure, the Q-value spectra are more “crowded”
than one would wish. Also, the use of singly charged projectiles limits the charge capture rate to a lower level than is desired. By using the EBIS, we will have fully stripped highly charged ions available, reducing both of these limitations. Thus, the future of MOTRIMS as a tool for measuring population dynamics in a variety of systems looks promising.

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