THE DIGITIZED ATOM AND OPTICAL PUMPING*

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ABSTRACT: We discuss how optical-pumping and double-resonance experiments on single atoms alter our experimental approach. Our knowledge of the atom can be in digital form; this allows an analysis quite different from experiments on ensembles of atoms.

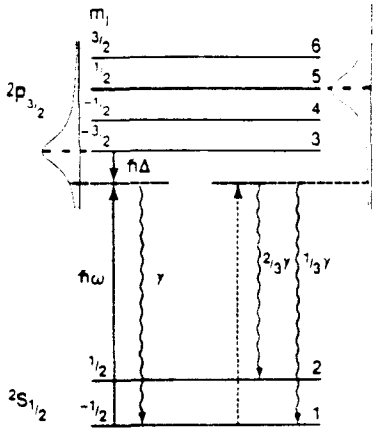
The methods of optical pumping and double resonance spectroscopy, as initiated by Kastler, Brossel, and their colleagues,[1] continue to be primary tools of experimental atomic physics. Until very recently, these techniques have been applied only to large ensembles of atoms. In these experiments, where the average behavior of many atoms is observed, the density (or "statistical") matrix formalism has been most useful in theoretically describing the experiments.

Technological advances have now allowed us to record the long term behavior of individual atoms. Recent interest has centered on the observation[2,3] and theoretical explanation[4] of "quantum jumps." The density matrix formalism can be used to describe quantum jumps, but the interpretation shifts from the average behavior of many atoms to the probability of a certain behavior (e.g., abrupt change in fluorescence intensity) occurring within an individual atom. In this note, we do not review theoretical explanations of quantum jumps,[4] rather, we briefly discuss how single atom experiments alter our experimental approach. Our knowledge of the atom can be in digital form; this allows an analysis quite different from experiments on ensembles of atoms.

$^{24}Mg^+$ IN A MAGNETIC FIELD[3]

A simple realization of single atom optical pumping is illustrated in Fig. 1. It employs a single radiation source which is tuned between a pair of Zeeman levels of an atom with a $^2S_{1/2}$ ground state and an excited $^2P_{3/2}$ state (for example, a neutral alkali atom or singly ionized alkaline earth atom such as $^{24}Mg^+$). The frequency ω of the radiation is tuned near the $S_{1/2}$, m_J=-1/2 $\leftrightarrow$ $P_{3/2}$, m_J=-3/2 transition frequency ω_0. The atom cycles nearly continuously between these levels since the dipole selection rules allow spontaneous decay only to the original ground level. A steady stream of fluorescence photons, which are readily detected, is emitted by the atom during this period. However, if the radiation is linearly polarized with the polarization direction perpendicular to the direction of the magnetic field, the $S_{1/2}$, m_J=-1/2 $\leftrightarrow$ $P_{3/2}$, m_J=1/2 transition is also allowed, although it is excited in the far wings of the curve for this resonance. This transition is indicated by the dashed arrow in Fig. 1. A spontaneous decay from the upper m_J=1/2 level can then leave the atom in the m_J=1/2 ground level. This optical pumping into the m_J=1/2 ground level takes the atom out of the m_J=-1/2 $\leftrightarrow$ m_J=-3/2

cycling loop, causing the emitted fluorescence to suddenly stop. The off-resonant $S_{1/2}$, m_J=+1/2 → $P_{3/2}$, m_J=-1/2 → $S_{1/2}$, m_J=-1/2 spontaneous Raman transition (not shown in Fig. 1) optically pumps the atom back to the m_J=-1/2 ground state where it will resume scattering.



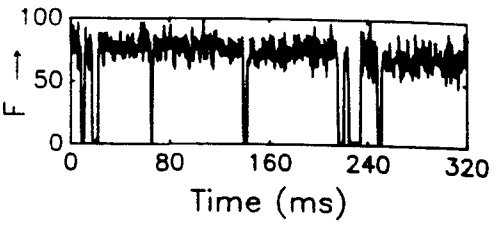


Figure 1. The energy level structure of the $^2S_{1/2}$ and $^2P_{3/2}$ states of an atom in a magnetic field (not to scale). A laser of frequency ω is assumed to be tuned near the $S_{1/2}$, m_J=-1/2 ↔ $P_{3/2}$, m_J=-3/2 transition frequency ω_0. The energy separation of adjacent $P_{3/2}(S_{1/2})$ sublevels is $\hbar\alpha/2$ ($3\hbar\alpha/4$) where $\alpha/2\pi$ = 52 GHz in the $^{24}Mg^+$ experiment. The indicated Lorentzian widths and detuning Δ, which are much less than α, have been exaggerated for clarity. At the right, a portion of the data, fluorescence (F) due to radiative decay from the $P_{3/2}$ state vs. time is shown. In this experiment, the dynamics of optical pumping between the m_J=-1/2 and m_J=+1/2 ground states is revealed in digital form as F switches on and off.

Single Atom vs Ensemble

The m_J=-1/2 ↔ m_J=-3/2 cycling transition serves only as a ground state monitor; optical pumping back and forth between ground state sublevels occurs due to the off resonant excitation described above. By examining the statistics of the discrete fluorescence level changes (quantum jumps) for a single ion, the pumping rates and average level populations can be derived. If we perform the same experiment on an ensemble of atoms, the discrete changes are averaged out and only a steady fluorescence is observed. In this case, we can obtain the pumping rates and average level populations by momentarily changing the average populations (for example, with another radiation source[5,6]) and monitoring the return of ensemble to its steady state. However, there is a simplicity to the single ion experiment where the dynamical evolution of the populations is revealed by the quantum jumps in the fluorescence intensity. (Also, the detection efficiency need not be held constant as in the ensemble experiments). In this particular experiment, our results agreed with theory[3] to within the measurement precision of 2%.

Hg^+ METASTABLE STATE LIFETIMES[7]

A single atom experiment which is related to the $^{24}Mg^+$ example is shown in Fig. 2. A single Hg^+ ion is illuminated by a laser tuned near the $^2S_{1/2} \rightarrow {}^2P_{1/2}$ transition. After excitation to the $^2P_{1/2}$ level, the ion usually decays back to the ground state but has a small probability of decaying to the $^2D_{3/2}$ level by emission of an 11 μm photon. The decay to the $^2D_{3/2}$ level is indicated by a sudden cessation of the 194 nm fluorescence as indicated in Fig. 2. From the $^2D_{3/2}$ level, the ion decays either to the ground state or to the metastable $^2D_{5/2}$ which eventually decays to the ground state. For brevity, we will only discuss the measurement of the lifetimes $\tau(^2D_{3/2})$, $\tau(^2D_{5/2})$, and the branching ratios, f_1 and f_2 shown below.

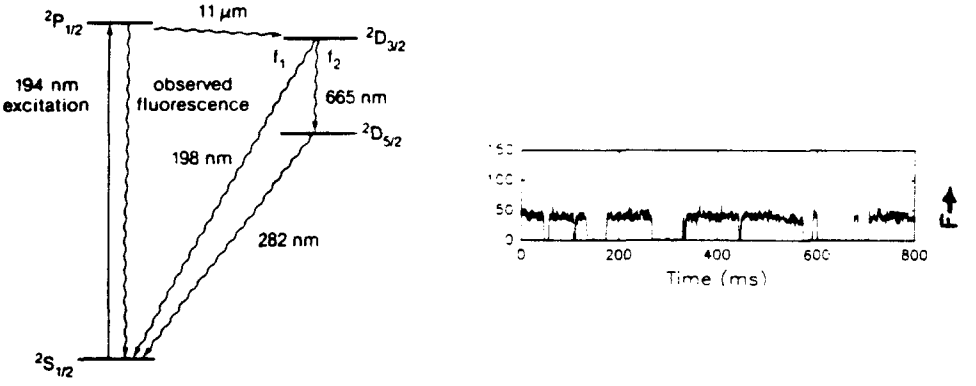


Figure 2. Diagram of the lowest four energy levels of Hg^+. The ion is continuously illuminated by radiation at 194 nm, and the fluorescence (F) at this wavelength is observed. The sudden cessations of fluorescence (at right) indicate the ion has decayed into the $^2D_{3/2}$ level.

Single Atom vs. Ensemble

By observing the statistics of fluorescence switching from a single ion (in particular, the distribution of fluorescence-off times) we can determine $\tau(^2D_{3/2})$, $\tau(^2D_{5/2})$, f_1, and f_2.[7] These quantities could be obtained from an experiment on an ensemble of atoms, by abruptly switching on the 194 nm laser and observing the transient behavior of the fluorescence. In the ensemble experiment, the 194 nm laser intensity and frequency and the fluorescence detection efficiency must be constant over the time the fluorescence is changing. In the single ion experiments we only require enough stability in these parameters that the jumps between "on" and "off" fluorescence be distinguishable. The single ion experiments also allow investigation of photon antibunching and sub-Poissonian statistics of the 11 μm emission.[8] This information is unique to single (or few) atom experiments and is not available in ensembles.

Hg^+ OPTICAL DOUBLE RESONANCE[9]

The narrow $^2S_{1/2} \rightarrow {}^2D_{5/2}$ quadrupole transition can be driven by a second laser. Transitions to the $^2D_{5/2}$ state can be detected by observing the abrupt cessation of the 194 nm fluorescence when theatom is excited to or "shelved" in[10] the $^2D_{5/2}$ level. In our experiment,[9] the lasers at 194 nm and 282 nm are alternately switched on and off (at approximately 20 ms intervals) in order to avoid a.c. Stark shifts on the quadrupole transition from the 194 nm radiation. When the 194 nm radiation is first turned back on, the absence of fluorescence indicates the ion has made the transition to the $^2D_{5/2}$ state, whereas the presence of fluorescence indicates the ion remained in the ground state after application of 282 nm radiation. We can supress false indications from the $^2P_{1/2} \rightarrow {}^2D_{3/2}$ (11 μm) emission by reducing the 194 nm laser power. The spectrum shown in Fig. 3 is the probability of seeing 194 nm fluorescence vs. the frequency of the 282 nm source.

Single Atom vs. Ensemble

As in the lifetime experiment above, the spectrum measurement is independent of 194 nm laser intensity and frequency or detection sensitivity fluctuations as long as the jumps are clearly distinguished. In an ensemble experiment,[11] or a single ion experiment where the detection sensitivity is not high enough to see quantum jumps,[6] the experiment is essentially the same but fluctuations in 194 nm intensity or fluorescence detection cause the signal to be more noisy. In the single ion experiment, the noise has reached the fundamental limit due to the quantum fluctuations in the $^2S_{1/2} \rightarrow {}^2D_{5/2}$ transition.[9]

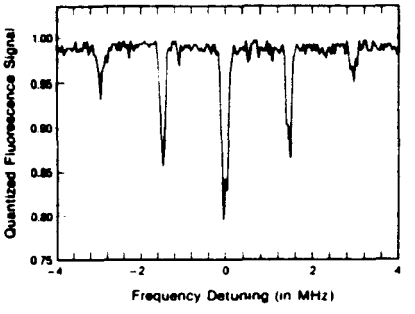


Figure 3. Quantized signal showing the electric-quadrupole-allowed $5d^{10}6s\ ^2S_{1/2}(m_J=-1/2)$ - $5d^96s^2\ ^2D_{5/2}(m_J=1/2)$ transition in a single, laser-cooled $^{198}Hg^+$ ion. On the horizontal axis is plotted the relative detuning from line center in frequency units at 282 nm. On the vertical axis is plotted the probability that the fluorescence from the $6s\ ^2S_{1/2}$ - $6p\ ^2P_{1/2}$ first resonance transition, excited by laser radiation at 194 nm, is on. Clearly resolved are the recoilless absorption resonance (carrier) and the Doppler sidebands due to the residual secular motion of the laser-cooled ion.

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