Baltimore /89

QUANTUM ELECTRONICS AND LASER SCIENCE CONFERENCE

1989 Technical Digest Series
Volume 12

Conference Edition

~~Summaries~~ of papers presented at the
Quantum Electronics and Laser Science
Conference
24–28 April 1989
Baltimore, Maryland

Sponsored by the
Optical Society of America
American Physical Society
Lasers and Electro-Optics Society of the Institute
of Electrical and Electronics Engineers

Optical Society of America
1816 Jefferson Place, N.W.
Washington, D.C. 20036
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reaction initiation. The motion toward the minimum of the S_1 potential energy surface apparently proceeds via two processes. Within 50 fs after absorption, equilibration of high frequency vibrational modes occurs. During this first reaction the molecule does not have time to move along the coordinates of the low-frequency reactive modes. The following slower reactive motion of the retinal is related to a 180-fs gain kinetic. In this process part of the isomerization of the retinal from the all-*trans* to the 13-*cis* configuration takes place, and the molecule arrives at the bottom of the S_1 potential energy surface in level S_1^{el}. The systems leave this area via internal conversion with a time constant of 500 fs. Two decay paths are used here. More than 60% of the molecules form the intermediate photoproduct *J*, while the rest returns to the original ground state of BR. Subsequently, the first ground-state reaction (in the active branch) proceeds with a time constant of 3 ps, leading to the intermediate *K* which is stable on the picosecond time scale. **(Invited paper, 25 min)**

1. J. Dobler, W. Zinth, W. Kaiser, and D. Oesterhelt, Chem. Phys. Lett. **144**, 215 (1988); W. Zinth, Naturwissenschaften **75**, 173 (1988).

Tuesday **MORNING**

25 April 1989 **TUCC**

HYATT REGENCY BALLROOM B

8:30 AM Trapping and Cooling: 1

David J. Wineland, U.S. National Institute of Standards & Technology, *Presider*

TUCC1 Order and chaos in laser cooled trapped ions

RALPH G. DEVOE, J. HOFFNAGLE, L. REYNA, R. G. BREWER, IBM Almaden Research Center, 650 Harry Rd., San Jose, CA 95120.

Several groups have recently used laser cooling to produce ordered arrays of ions (ion crystals) in rf quadrupole traps. We discuss two physical mechanisms which underlie and explain the behavior of ion crystals. First, we show that deterministic chaos can be responsible for phase transitions between the ordered (crystal) state and the disordered (cloud) state.[1] The crystal—cloud transition is explained as an order—chaos transition. Chaos arises from the nonlinear interaction of Coulomb repulsion with the oscillating trap potential. Numerical integration of the resulting Mathieu equation with a Coulomb term shows an abrupt transition to a chaotic state as the trap voltage is raised above a threshold value. Trajectories in the disordered state display a positive Lyapunov exponent and satisfy three other tests for chaos. Experiments on two trapped Ba^+ ions show a crystal—cloud transition at the predicted value of trap voltage.

Second, we discuss an unexpected laser cooling instability that limits the size of ion crystals in rf traps and can also initiate an order—chaos transition. Previous laser cooling theories assumed a static harmonic trap potential and ignored the micromotion arising from the oscillating rf trap field. This is valid for single ions but not for crystals, where micromotion typically exceeds several wavelengths of light. The resulting Doppler shifts frequency modulate the cooling radiation and can transform laser cooling into laser heating. This mechanism explains the complex dependence of the phase transitions on trap parameters as well as the failure to crystallize more than 100 ions in rf traps. It also permits order—chaos transitions to occur over a broad range of trap parameters.

(Invited paper, 25 min)

1. J. Hoffnagle, R. G. DeVoe, L. Reyna, and R. G. Brewer, Phys. Rev. Lett. **61**, 255 (1988).

TUCC2 Radiofrequency spectroscopy of trapped neutral atoms

D. E. PRITCHARD, KRISTIAN HELMERSON, MIN XIAO, KE-XUN SUN, Massachusetts Institute of Technology, Cambridge, MA 02139.

We have recently demonstrated rf resonance experiments on Na atoms contained in a magnetic trap.[1,2] Sequential rf pulses with appropriate frequencies were applied to a sample of $\sim 2 \times 10^{10}$ Na atoms (confined in an $\sim 100\text{-cm}^3$ volume) to transfer population between the four trapped magnetic substates. The relative populations of the magnetic substates were detected by sending a weak narrowly collimated probe laser beam along the axis of the trap and monitoring the induced fluorescence with a photodiode (placed along the wall of the trap enclosure) located at the longitudinal magnetic field minimum. Hence we are sensitive only to light emanating from the small fraction of atoms near the magnetic field minimum.

To diagnose the energy distribution of the trapped atoms, we measured the rf resonance curve for the $|F = 2, M = 2\rangle$ to $|F = 2, M = 1\rangle$ transition by measuring the relative peak heights in the fluoresence spectrum for the two states as a function of the frequency of the applied rf pulse. Figure 1 is such an rf resonance curve for both hot atoms after the initial loading of the trap and cold atoms after the application of 1-D Doppler cooling. Doppler cooling of the sample was achieved with a standing wave laser beam, detuned ~80 MHz red of the trapped atom transition, applied for 60 s. It was necessary to reduce the intensity of the standing wave to $\sim 10^{-3}$ saturation intensity to make the transverse heating rate slow compared with the coupling rate of transverse to longitudinal motion provided by the trap.

The energy difference between the two levels increases monotonically with the magnetic field $[d(E/h)/dB \sim 60\ \text{kHz/G}]$. Hence the rf frequency is resonant only with atoms at one value of the magnetic field. The applied rf pulses were of sufficient power and duration (number of Rabi flops: $5\ \text{s} \times \Omega_R/2\pi \sim 1 \times 10^4$) so that equilibrium was reached in the transfer of population between the two states. The height of the resonance curve at each frequency is, therefore, determined by the fraction of atoms energetic enough to reach the corresponding magnetic field, and the line shape of the high-frequency tail, in the collisionless regime, becomes a direct measure of the energy distribution of the atoms in the trap. Since our population ratio measurements of atomic density occur in a small-volume element located at the magnetic field minimum of the trap, there is no coordinate-space density of states dependence, making the shape of the spectrum a direct measure of the energy distribution, independent of the trap potential. Thus it is possible to extract the energy distribution of the trapped atoms from the resonance curves of Fig. 1. Figure 2 is a plot of the logarithm of the energy distribution as a function of energy for the data of Fig. 1. If the energy distribution were a field-mean distribution, we could expect a straight line. The linear fits to [illegible] suggest that the hot atom distribution is a truncated Boltzmann distribution with a temperature of 60^{+40}_{-20} mK, while the cold atom distribution is approximately Boltzmann at higher energies with a temperature of ~2 mK. **(Invited paper, 25 min)**

1. V. S. Bagnato, G. P. Lafyatis, A. G. Martin, E. L. Raab, R. N. Ahmad-Bitar, and D. E. Pritchard, Phys. Rev. Lett. **58**, 2194 (1987).
2. A. G. Martin, K. Helmerson, V. S. Bagnato, G. P. Lafyatis, and D. E. Pritchard, Phys. Rev. Lett. **61**, 2431 (1988).

TUCC3 Measurement of fluorescence along the deceleration path of an atomic beam

V. S. BAGNATO, S. C. ZILIO, U. Sao Paulo, Institute of Physics & Chemistry of San Carlos, Department of Physics & Science of Materials, C. P. 369, 13560 Sao Carlos, SP, Brazil.

The production of decelerated atomic beams of alkalines is now routine in several laboratories. Usually, the observation is carried out by using a second weak laser to perform a velocity analysis on the beam after the deceleration process. We report the observation of the laser cooling process in an atomic beam of sodium by monitoring the fluorescence of the atoms along the deceleration path,[1] using for this purpose a single laser to both decelerate and probe the cooled atoms. A simple theoretical model describes very well the experimental results.

An experimental setup somewhat similar to the one presented in Refs. 2 and 3 is schematically shown in Fig. 1. The 130-cm long solenoid has an axial magnetic field of $B = B_b + B_0\sqrt{1 - \beta(z - z_0)}$, where $B_b \sim 300$ G, $B_0 \sim 1000$ G, and $\beta = 0.01\ \text{cm}^{-1}$. The laser beam is provided by a ring dye laser with the light circularly polarized and tuned to a σ^+ transition from the $F = 2$, $M_F = 2$ sublevel of the $^3S_{1/2}$ ground state to the $F = 3$, $M_F = 3$ sublevel of the $^3P_{3/2}$ excited state.

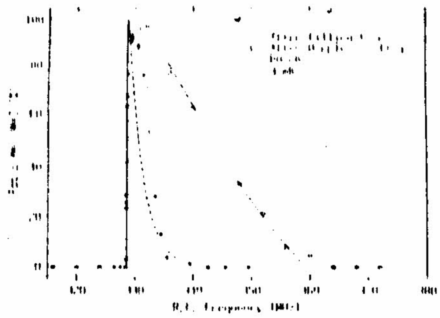
A set of Si photodiodes is located around the atomic beam which runs inside a glass tube concentric with the solenoid. This detector assembly can be moved along the whole extension of the magnetic field and collects light coming from the fluorescence of the atoms during the deceleration process. A small portion of the laser light crosses the atomic beam at a right angle before it enters the solenoid, and the fluorescence arising from this interaction is used as a reference for the frequency corresponding to $v = 0$ and $B = 0$.

Fluorescence as a function of the frequency is shown in Fig. 2 for different positions of the detector assembly. We arbitrarily chose the $z = 0$ position at the solenoid entrance close to the sodium source. The arrows indicate the frequencies corresponding to $v = 0$ for atoms subjected to the magnetic field at that z position. The first thing to be pointed out is that as z increases the curves shift toward the $v = 0$ frequency. This is evidence that the beam has been decelerated. The shape of these curves does not reflect the velocity distribution directly.

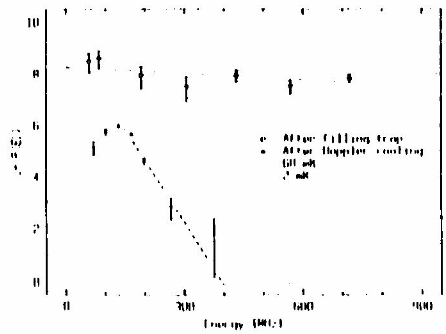
A simple analytical model gives the fluorescence signal as

$$S(z) = \frac{S_0}{B(z) - B_b}\left\{\exp[-v^2(z)/\alpha^2]\left[\frac{v^2(z)}{\alpha^2} + 1\right] - \exp(-v_0^2/\alpha^2)\left(\frac{v_0^2}{\alpha^2} + 1\right)\right\},$$

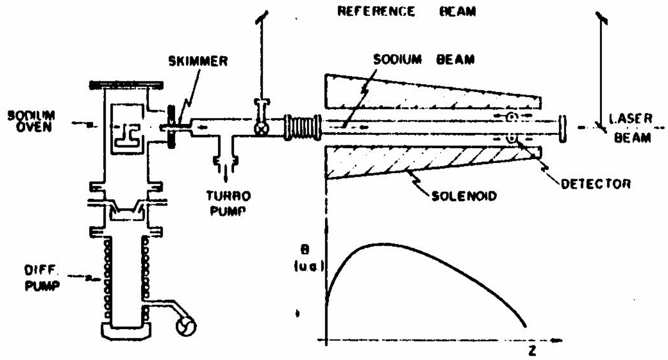
where $\alpha = \sqrt{2k_BT/M}$, and v_0 is the maximum velocity which can be decelerated for a given value of B_0. A plot of $S(z)$ as a function of the laser detuning gives a curve with a shape similar to the one in



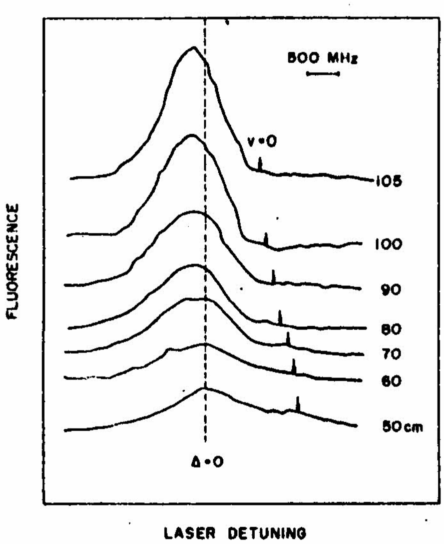
TUCC2 Fig. 1. Radio-frequency resonance curve for atoms after initial loading of the trap (open circles) and application of Doppler cooling (solid circles). The dashed and solid lines are theoretical fits of 3 and 60 mK, respectively.



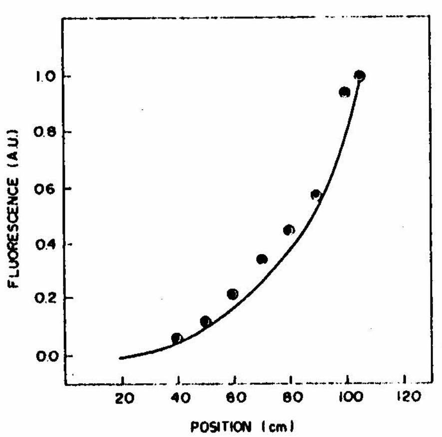
TUCC2 Fig. 2. Logarithm of the energy distribution $P(E)$ vs energy. The solid and dashed lines represent Boltzmann distributions of 60 and 2 mK, respectively.



TUCC3 Fig. 1. Schematic diagram of the apparatus used in the laser cooling experiment with a plot of $B(z)$ vs z.



TUCC3 Fig. 2. Fluorescence intensity as a function of laser detuning for several positions of the detector assembly.



TUCC3 Fig. 3. Fluorescence intensity of the detector assembly position for $\Delta = 0$. The background scattered light is already subtracted.

Fig. 2. On the other hand, for $\Delta = 0$ we can plot the amplitude of the fluorescence as a function of the position. This is shown in Fig. 3 together with the experimental points. The good agreement between the model and experimental points means that the main physical aspects have been taken into account in the model. The signal observed is consistent with an atomic flux of 5×10^{13} atoms/s in contrast with the initial flux estimated to be of the order of 10^{15} atoms/s. (12 min)

1. A similar procedure to observe deceleration of an atomic beam has also been used at MIT, and a paper by V. S. Bagnato, G. Lafyatis, A. Martin, K. Helmerson, and D. Pritchard is being prepared for publication.

2. W. P. Phillips and H. Metcalf, Phys. Rev. Lett. 48, 596 (1982).
3. J. V. Prodan, W. Phillips, and H. Metcalf, Phys. Rev. Lett. 49, 1149 (1982).

TUCC4 Application of quantum jumps: the accurate determination of quenching rates for a single trapped barium ion

A. A. MADEJ, J. D. SANKEY, National Research Council of Canada, Time & Length Standards, Ottawa, Ont. K1A 0R6, Canada.

Recent experiments on single trapped ions[1-3] show the quantum nature of the system through the observation of what is popularly called quantum jumps. By monitoring the emitted light from the trapped ion subject to excitation by laser radiation, the presence or absence of fluorescence indicates whether the ion exists in a nonfluorescing metastable state. We have observed the quenching of the $^2D_{5/2}$ state in Ba^+ under the influence of a number of atomic and molecular gases at ultrahigh vacuum (10^{-6}–10^{-8} Pa) by the observation of changes in the quantum jump statistics. A number of gases were studied which typically make up the residual background gas environment of ultrahigh-vacuum systems. In our experiment, accurate residual gas analysis and absolute quenching gas pressure were provided with calibrated mass spectrometer and ion gauge measurements. This enabled us to include the effects of residual impurity gases which can severely alter the measurements of a particular quenching gas. Also, the arrangement of an otherwise unperturbed single trapped laser cooled ion ($T < 1$ K) suffering collisions from a well-characterized background gas provides an experimentally simple and direct system for analysis of collision events. The present work also allows us the ability to characterize completely the environment of the single trapped ion in the background gas which exists in our vacuum chamber. This is important in our work toward establishing an accurate error analysis in the use of a single trapped ion as an optical frequency standard.[4] (12 min)

1. W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. 56, 2797 (1986).
2. Th. Sauter, W. Neuhauser, R. Blatt, and P. E. Toschek, Phys. Rev. Lett. 57, 1696 (1986).
3. J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. 57, 1699 (1986).
4. D. J. Wineland, Science 226, 395 (1984).

Tuesday MORNING
25 April 1989 JC

HYATT REGENCY BALLROOM C/D

10:30 AM Joint CLEO/QELS Symposium on High Field Effects: 2

S. E. Harris, Stanford University, Presider

JC1 All high intensity multiphoton ionization is necessarily resonant

RICHARD R. FREEMAN, AT&T Bell Laboratories,

At high intensities, the excited states of atoms are shifted by the ponderomotive potential of the laser field and come into resonance with some harmonic of the laser. The energy spectra of the photoelectrons and the ionic spatial distributions are dominated by three resonances. (Invited paper, 25 min)

JC2 Above threshold ionization: a controlled plasma heating mechanism

PAUL B. CORKUM, N. H. BURNETT, F. BRUNEL, National Research Council of Canada, Division of Physics, Ottawa, ON K1A 0R6, Canada.

Understanding multiphoton ionization of atoms or molecules is of fundamental importance in atomic and molecular physics. However, it is rarely noted that it is equally important for plasma physics. Laboratory plasmas are normally ionized and heated by electron collisions. Plasmas produced by photons will have very different characteristics.

It is easiest to understand multiphoton ionization in the long wavelength limit because in this limit classical physics can be applied to much of the problem. Consider an electron released at time t' from rest into a strong laser field. The electron will experience a force which in the nonrelativistic limit is proportional to the electric field $\mathbf{E} = E_0 \cos(\omega t)\mathbf{e}_x + \alpha E_0 \sin(\omega t)\mathbf{e}_y$, where $\alpha < 1$ allows for arbitrary polarization.

The electron motion in the field of the laser is given by

$v_x = v_0 \sin(\omega t) + v_{0x}$, $v_y = -(\alpha v_0) \cos(\omega t) + v_{0y}$, where $v_0 = qE_0/m\omega$ and v_{0x} and v_{0y} can be evaluated from the initial conditions. $v_{0x} = -v_0 \sin(\omega t')$ and $v_{0y} = \alpha v_0 \cos(\omega t')$ is the drift energy of the electrons produced by multiphoton ionization. The probability of ionization as a function of time can be calculated in the quasistatic limit using dc tunneling theory.[1] Thus tunneling determines the distribution of drift velocities.

We show that experiment confirms these predictions. In particular, using a 10-μm pulse to ionize xenon we demonstrate that by the simple procedure of changing the laser polarization, the electron temperature (drift energy) can be determined anywhere between 50 and 800 eV. If we were to change the ionizing laser wavelength, the drift velocity would change even further. Finally, the ionization potential of the atom or ion can also be considered a variable thus giving even greater flexibility in selecting electron temperatures.

This control over the electron temperature of an underdense inertially confined plasma has many important implications. Plasmas produced by multiphoton ionization are not in thermal equilibrium. Using practical lasers, plasmas can be produced where the electron temperature is much less than the ionization potential of the ion from which the last electron was extracted. Such a situation should lead to transient (x-ray) gain during the recombination phase of the plasma. Even ground state lasers are predicted. In the other extreme, energetic electrons produced using long wavelength lasers and high ionization potential ions will represent an extremely high specific power deposition.

Before concluding, we point out other implications of multiphoton ionization for plasma physics. (1) New nonlinear phenomena are introduced by the process of plasma formation. (2) The presence of even relatively low levels of radiation can inhibit the recombination process. (3) It should be possible to produce energetic ions by removing a number of molecular electrons in rapid succession. This process, known as a Coulomb explosion,[2] will require pulses with a duration of the order of the vibrational time of the molecule. Such rapid electron removal is possible in atomic systems[3] and should be possible with molecules. (4) Using large molecules or clusters, microplasmas can be produced which will exhibit some of the characteristics of laser-solid target interactions and some of the characteristics of gaseous targets. (Invited paper, 25 min)

1. L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon, New York, 1965), p 276.
2. K. Codling, L. J. Frasinki, P. Hathorly, and J. R. M. Barr, J. Phys. B 20, L525 (1987).
3. S. L. Chin, C. Rolland, P. B. Corkum, and Paul Kelly, Phys. Rev. Lett. 61, 153 (1988).

JC3 Molecular bonds in intense laser fields

P. H. BUCKSBAUM, A. ZAVRIYEV, D. W. SCHUMACHER, AT&T Bell Laboratories, Murray Hill, NJ 07974.

Above-threshold ionization (ATI) and dissociation of molecular hydrogen in intense laser beams show the interplay between electrostatic and radiation fields in molecules at intensities above 10^{13} W/cm². In these experiments, H_2 was irradiated by intense 100-ps light pulses at 532, 355, or 1064 nm. Both electron and ion energy spectra were obtained using a field-free angle-resolving time-of-flight spectrometer. Angular distributions were also measured for electrons and protons. Considerable precautions were taken to eliminate space charge distortion of the spectra.

The photoelectron spectra at 355 and 532 nm contain ATI peaks that display the vibrational fine structure of the H_2^+ ground state (Figs. 1 and 2). This suggests that photoionization occurs prior to dissociation and that the H_2^+ ion remains stable for at least several vibrational periods prior to double ionization or dissociation. In addition, the ion spectra do not contain the high energy component that would accompany direct two-electron ionization from the ground state followed by Coulomb repulsion. These observations lead us to conclude that direct double ionization does not occur for these intensities and wavelengths.

The vibrational levels in the visible and UV ATI spectra change their relative strengths and angular distributions with pulse energy. As the peak intensity increases, the vibrational levels gradually broaden and ultimately merge into a broad peak. At the same time, the amount of above-threshold ionization increases with intensity as it does for atoms.

The electron spectra at 1064 nm do not show a vibrational structure. Neither their shape nor intensity dependence has features commonly attributed to ATI (Fig. 3). Instead, the electrons form a continuous spectrum with a maximum near 10 eV. This nearly featureless distribution can extend to more than 70 eV. The high energy tail increases about linearly with intensity.

(Invited paper, 25 min)

Campo	Dado
*****	Documento 1 de 1
No. Registro	000802289
Tipo de material	TRABALHO DE EVENTO-RESUMO - INTERNACIONAL
Entrada Principal	Bagnato, Vanderlei Salvador
Título	Measurement of fluorescence along the deceleration path of an atomic beam.
Imprenta	Washington : Optical Society of America, 1989.
Descrição	ref.tucc3.
Autor Secundário	Zílio, Sérgio Carlos
Autor Secundário	Quantum Electronics and Laser Science Conference (1989 Baltimore)
Fonte	Abstracts, Washington : Optical Society of America, 1989
Unidade USP	IFQSC-F -- INST DE FÍSICA DE SÃO CARLOS
Localização	IFSC PROD001241