Ultracold neutral plasmas

S. L. Rolston and J. L. Roberts
Atomic Physics Division
National Institute of Standards and Technology
Gaithersburg, MD 20899-8424

Abstract

Photoionization of samples of laser-cooled atoms has allowed the creation of ultracold neutral plasmas, accessing an unexplored plasma parameter regime. Among the phenomena that have been observed are a rapid expansion of the unconfined plasma, and recombination of the plasma into Rydberg atoms, even at very low plasma densities. The expanding plasma is a complex dynamic system, with adiabatic cooling, evaporative cooling, and recombination heating all occurring simultaneously. Current experiments seek to determine the temperature evolution of the plasma that results from these competing processes.

1 Introduction

Neutral plasma physics has studied an immense range of temperatures, from 10^{16} K in the magnetosphere of a pulsar to 300 K in the Earth's ionosphere. Lower temperatures are both difficult to achieve and interesting for many reasons. For instance, as the temperature is lowered, the relevant collisional mechanisms can change. In particular, three-body recombination, which scales as $\mathrm{T}^{-9/2}$, should become dominant. Another interesting regime can be accessed by low temperature plasmas if the Coulomb energy between nearest neighbors in the plasma becomes comparable to or larger than the thermal energy. In that case, correlations become important and the plasma enters a strongly coupled regime where many of the standard classical plasma physics assumptions become invalid. The strong-coupling regime for electrons is characterized by the dimensionless Coulomb coupling parameter

$$\Gamma = (\frac{e^2}{4\pi\epsilon_0 a})/k_B T_e \tag{1}$$

where $a = (4\pi n/3)^{-1/3}$ is the Wigner-Seitz radius, n is the electron density, and T_e is the electron temperature. When $\Gamma \geq 1$ correlations in the plasma start to become important and the plasma begins to enter the strongly coupled regime. At high enough Γ, liquid and solid states are possible. In nature, such plasmas are thought to exist at the center of massive planets and white dwarfs. They are also relevant for inertial confinement fusion.

Using atomic physics techniques, we are able to produce ultracold neutral plasmas, which have initial temperatures as low as a few Kelvin. These are produced by photoionizing a sample of laser-cooled atoms. The excess energy above the ionization limit of the photoionizing photon provides the energy to the plasma. Since the frequency of the photoionizing laser is tunable, the energy imparted to the plasma can be controlled. The use of laser-cooled atoms means there is essentially no contribution to the plasma energy from the thermal energy of the initial neutral sample of atoms. The experiments performed at NIST that we describe here used xenon atoms, but we expect that experiments performed with other atoms should yield similar results since the particular properties of xenon are not believed to play a role in the plasma creation or behavior. We can create plasmas with initial densities of 10^9 cm^{-3} and temperatures varying from 1-1000K, depending on the energy of the ionizing photon. Whether or not these plasmas enter the strongly coupled regime depends on their evolution and remains an open question.

Our first observations of the creation of an ultracold neutral plasma [1] in 1999 were done in a theoretical vacuum, and thus represented a chance to do exploratory physics. We verified that we could indeed create a plasma (there was some speculation that three-body recombination would be so rapid that a plasma could never be formed). We measured the existence of plasma oscillations and used them to extract the density, which was found to decrease with time. This decrease in density was expected, because the plasma is created with no confining fields and so is free to expand. Experiments revealed that the expansion velocity is in large part dominated by the thermal pressure the electrons exert on the ions. We have measured the production of Rydberg atoms, presumably due in part at least to three-body recombination, although there are interesting puzzles that remain. Our group as well as others has observed that a dense gas of cold Rydberg atoms can spontaneously evolve into a cold plasma. This experimental effort has spurred a number of theoretical groups to get involved, with some success as discussed below. But there remain many interesting questions in this 3-year old field.

2 Creation of ultracold neutral plasmas

Our plasmas are created by the photoionization of a cloud of metastable xenon atoms that are confined and cooled by a magneto-optical trap (MOT) to typical densities of 10^9 cm^{-3} and temperatures near 10 μK. The choice of metastable xenon was determined by the fact that the system was available for experimentation, and the laser wavelength for photoionization was convenient. We use a two-step photoionization process, using a photon (882 nm) tuned to the $6s[3/2]_2 \rightarrow 6p[5/2]_3$ transition (also used for laser cooling) and a photon generated by a pulsed (10 ns) dye laser (514 nm) from the $6p[5/2]_3$ state to the continuum. The amount of photoionization is controlled by the intensity of the pulsed laser, and can be as high as 35%. The vacuum chamber is a standard laser-cooling system, with the addition of fine wire mesh grids surrounding the MOT region. The grids are used to apply electric fields to the plasma and to extract electron from the plasma. Once extracted, the electrons are accelerated by a second grid onto a microchannel plate (MCP) detector at the bottom of the vacuum apparatus. The signal is large enough so that the MCP is operated in analog current mode. We found that the detector signal could easily become saturated if the flux of arriving electrons was high enough, and so care was taken to assess and minimize the detector saturation in each experiment. In addition to the electrons, it is also possible to detect the ions in the plasma by reversing the field directions. However, the short time-of-flight of electrons make them more suitable for the experiments discussed here, especially when detecting plasma evolution on the μs timescale.

The plasma formation proceeds as follows. Immediately after the photoionization, the electrons are monoenergetic, with a kinetic energy given by the excess energy of the ionizing photon above the ionization limit. The electrons carry off almost all of this excess energy and the ions receive very little of it. This is due to the large ratio in masses between the ion and electrons. The ions will only gain 4×10^{-6} of the initial energy, remaining in the sub-mK temperature range. They can be considered to be essentially stationary at short timescales. As the electrons thermalize (ns timescale)[2], a few of them leave the cloud. This creates a residual positively charged cloud, which begins to act as an attractive potential for the remaining electrons. This process will continue until the potential is as deep as the temperature of the electrons, at which point they are trapped, forming a neutral plasma. This has been verified by studying the number of trapped electrons as a function of the number of ions formed in the ionization process. A simple integration of Newton's equations for a set of charged shells found excellent agreement with the data presented in Ref. [1]. Recent state-of-the-art molecular dynamics simulations carried out at Los Alamos [3] and the University of San Diego [4] confirm this interpretation. Although there must be some overall positive charge to create the plasma, we can operate under conditions where the neutrality is greater than 96%. In reality, the density of ions and electrons throughout most of the plasma are more closely matched than that just the ratios of their total numbers would suggest. This is because the electrons distribution will respond to the ion distribution to attempt to neutralize the ion cloud's electric field, creating an even more nearly neutral plasma in the central region. At the edge one runs out of electrons to neutralize the ions, and so it is expected that there will be positively charged outer shell of ions.

3 Plasma expansion

While the electrons in the plasma are confined by the field produced by the ions, the neutral plasma on the whole is unconfined and will expand. A naive estimate for the expansion might be that it would expand at the residual thermal velocity the ions possess immediately after the plasma forms. This would lead to a ms timescale for expansion. Due to the mass difference between the hotter electrons and the ions, thermalization via elastic collisions between the two components is inefficient and would occur over a several ms timescale — hence the residual thermal velocity of the ions at the time of plasma creation is appropriate for this nave estimate. In ref.[5] the expansion time was found to be on the μs time scale instead. The expansion rate of the plasma estimated from the residual charge imbalance between ions and electrons could also not account for the observed expansion rate.

This expansion rate was measured by observing the response of the plasma to an RF electric field. The fundamental collective mode of a plasma, the plasma (Langmuir) oscillation, is a mode in which the electrons oscillate around their equilibrium positions and the ions remain essentially stationary. The oscillation frequency of this collective mode is a function of the electron density and is independent of temperature. When the density of the plasma was such that a large fraction of the plasma could be excited by the RF coupling to this plasma mode, energy was pumped in and electrons were boiled off. These electrons then appeared as a peak in detected electron current at a certain time after plasma creation (see Fig. 1). By interpreting the time of the peak of this response as the time when the plasma had the average density corresponding to the density that would be resonant with the applied RF drive, it is possible to extract the density vs. expansion time, i.e. the plasma expansion rate (see Fig. 2). Ref [5] found that at higher initial energies ($E_e/k_b \geq 50K$), the plasma expanded with a velocity proportional to $\sqrt{kT_e/m_i}$, namely proportional to the *electron* temperature and the *ion* mass. This can be understood in terms of a pressure exerted by the electrons on the ions. If we envision electrons oscillating in the Coulomb potential created by the excess ions, whenever an electron is turned around by the potential it exerts a force on the ions, leading to an outward expansion of the ions. It should be emphasized that this increase in ion velocity does not correspond to an increase in the ion temperature, as it represents a coherent expansion. The ion temperature (characterized by the *random* thermal motion) can still be quite cold. This rapid expansion has been successfully modeled with a Fokker-Planck treatment [6] as well as molecular dynamics simulations [3].

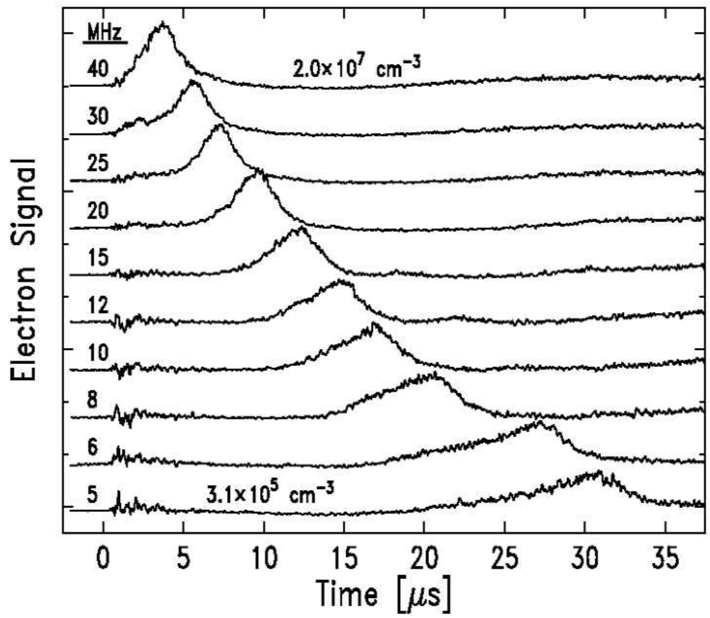

Figure 1: Plasma response to applied RF fields [5]. Each trace in the figure represents a different measurement; the different traces are offset for clarity. The applied RF frequency (MHz) is shown on the left. A background subtraction has been performed on the data to remove the signal due to electrons that normally leak out of the plasma (i.e. without the RF being applied).

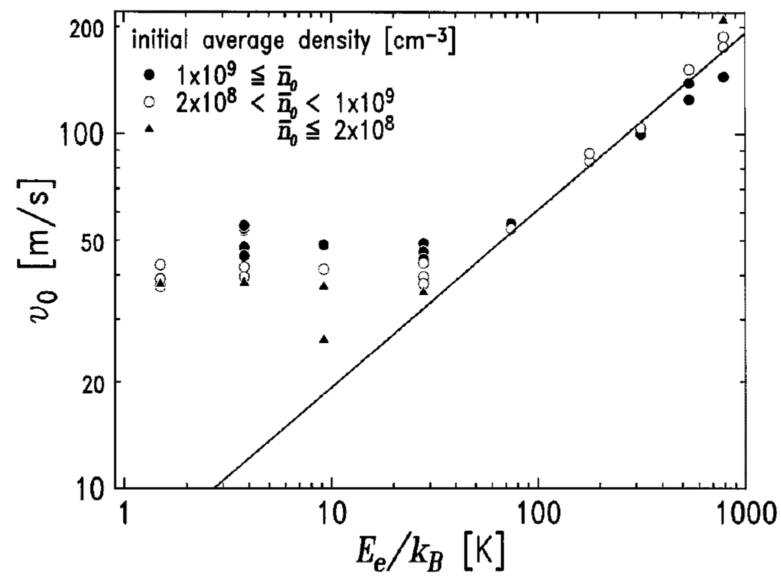

Figure 2: The asymptotic expansion velocity of the plasma as a function of the initial energy imparted to the electrons by the photoionization [5]. The straight line is the expansion velocity that would be expected assuming a dynamic equilibrium is reached between the expansion rate and the electron temperature.

4 Rydberg formation

In [5] it was found that there was an anomalous excess expansion velocity at low temperatures that could not be explained by electron pressure (see Fig. 2). This might seem troubling because it would imply there was more energy in expansion than had been supplied by the photons doing the ionization. Of course energy conservation must hold, which means there must also be present a source of negative energy, i.e. bound atoms. We performed an experiment [7] to look for the formation of Rydberg atoms. These Rydberg atoms were detected by using selective field ionization after the plasma was created (i.e. the application of a large electric field to field ionize the atoms). We found as much as 30% of the plasma recombines into Rydberg atoms. The binding energy of the Rydberg atoms accounted for the excess expansion energy, although it is difficult to do this measurement to better than a factor of two.

An examination of the possible recombination mechanisms rules out all processes at low temperatures except three-body recombination, which is predicted to scale as $T^{-9/2}$ [8] and could be quite large at low temperatures. Three-body recombination is an inelastic collision where two electrons and an ion [9] come in and a Rydberg atom and electron go out. Energy and momentum balance in the collision is maintained as the third-body electron carries off an energy roughly equal to the binding energy of the atom formed. Through elastic collisions, this third-body electron will quickly come into thermal equilibrium with the other electrons in the plasma, and so the binding energy released in the Rydberg formation will increase the temperature of the electrons in the plasma.

Measurements of the principal quantum number distribution found more tightly bound atoms at higher densities and lower temperatures. A simple application of the three-body recombination rates would have trouble explaining these trends. Not only is no density dependence predicted, the simple rate equations would imply that the lower the temperature the more loosely bound the atoms formed, in contradiction with the observations. Subsequently a theoretical treatment [6] explained this apparent discrepancy by including the effects of electron-Rydberg collisions. These collisions drive atoms to more deeply bound levels, and are more effective at higher density. Molecular dynamics simulations [3, 10] also find Rydberg atom formation.

Although the theory in Ref. [6] seemed to successfully explain the data of Ref. [7], there remain some troubling but interesting discrepancies. Experimentally it is found that the Rydberg production rates are roughly constant over a 100 μs timescale, where the plasma density has fallen by 4 orders of magnitude. The theory of ref. [6] was essentially done for short times (as were the molecular dynamics simulations), and predicts that the Coulomb coupling constant approaches a constant value, i.e. that $n^{1/3}T^{-1} = const$. Since the density falls as t^{-3}, this implies that $T \propto t^{-1}$ and therefore the three-body rate will fall as $t^{-3/2}$, in contradiction with the observed constant Rydberg production rate. Yet the theory clearly has some relevance as it is rather successful at predicting the expansion velocity. Another puzzle independent of theory is that the expansion velocity appears to asymptote in 10 μs, while the only a small fraction of the Rydberg atoms have been produced. So how can Rydberg production account for the energy balance when most of the Rydbergs have yet to be formed?

5 Temperature measurements

The expanding ultracold neutral plasma is a complex dynamical system, and its electron temperature vs. time behavior is influenced by a number of factors. One of the factors that needs to be taken into account is that when the plasma is formed the resulting initial energy per electron may in fact be greater than what would be suggested from the photoionization photon energy alone. The molecular dynamics simulations [3, 4, 10] show that very quickly after plasma creation the plasma will convert any correlation energy into electron thermal energy, rapidly driving Γ toward 1, if it was initially $\gg 1$. (Because there were no correlations in the neutral atom cloud from which the plasma was formed, the creation of the plasma at high Γ results in a state without the appropriate correlations that would exist in thermal equilibrium at high Γ). This mechanism would place a lower limit on the initial temperature of the plasma.

Once the plasma is created at a particular temperature, that temperature is not expected to remain constant as the dynamics during its expansion will also have a great impact. As the plasma expands, the electron thermal energy is converted into ion expansion velocity, resulting in strong adiabatic cooling of the electron cloud. In addition, we observe electrons leaking out of the plasma after the initial burst that escapes when the initial confinement and charge imbalance occurs during formation. These electrons that slowly leak out of the plasma are presumably from the high-energy tail of the Boltzmann distribution, and so their escape results in evaporative cooling.

These cooling mechanisms are in competition with the heating due to three-body recombination. Because of the strong inverse T dependence, we would expect the recombination will try to act as a temperature moderator in any circumstance where a significant number of electrons recombine. Any cold plasma will have a fast recombination rate that will then in turn heat the plasma. Any hot plasma can cool (through adiabatic expansion or evaporation) until the recombination rate picks up, adding enough heat to slow the cooling. Whether or not such temperature regulation is occurring is an area of current experimental interest.

How three-body recombination proceeds when the temperature gets quite low is an open question[11]. The $T^{-9/2}$ scaling is a classical plasma result, which is not expected to hold if the plasma begins to become strongly coupled. Whether or not Γ can actually increase above 1 into the strong coupling regime is tied into the dynamics. If there were no heating effects, adiabatic cooling alone would have Γ increase as the plasma expands.

Measuring the temperature of such an ultracold plasma is a challenge, as its transient nature and the low temperatures means that invasive physical probes such as used with traditional plasmas will not work. One possible avenue that has been used in plasma physics is to measure the ion acoustic velocity which is proportional to $T_e^{1/2}$. We have seen evidence for ion acoustic waves under various parameters, where we observe multiple peaks in the response of the plasma to the RF field that excites plasma oscillations. We interpret these peaks as ripples in the density profile due to the excitation of ion acoustic waves at the plasma formation. Such waves have also been seen in theory simulations [6]. The problem with using them as a measure of the temperature is that these waves are traveling on a expanding density profile that is expanding with essentially the same velocity as the acoustic wave. This along with the inhomogeneous density profile makes any simple analysis impossible.

We have been developing a different technique to measure the electron temperature, by observing the tail of the Boltzmann distribution of electron velocities in the plasma. At a variable time after the formation of the plasma, we suddenly turn on a weak electric field (10-30 mV/cm). The addition of this field will lower the barrier of the Coulomb potential confining the electrons, allowing any electrons with energies above the barrier to escape and be detected. (This is similar to selective field ionization used for Rydberg atom detection.) We record the number of electrons released as a function of the size of the spilling field.

In order to determine the temperature of the plasma from this data, we need to be able to relate the number of electrons spilled out as a function of applied electric field to the temperature of the plasma. The spilling process is modeled in two steps. First, a self-consistent electron distribution in thermal equilibrium is computed for a given ion number, electron number, plasma spatial size, and electron temperature. Once this is done, an external field is added to the potential and the amount of lowering in the Coulomb barrier is computed. All of the electrons with energy greater than this barrier height are then presumed to escape and the fraction spilled out as a function of applied field is thus determined. In order to get agreement between the data and the prediction, it was necessary to include the screening response of the electrons to the applied external field.

We generally find that the fraction of electrons spilled is a linear function of the applied electric field. A slope and intercept can therefore characterize the spilling for a given set of experimental conditions. Initially, we had planned on using the slope to determine the temperature. However, studies of the plasma showed that deliberate increases in the plasma temperature did not change the slope an appreciable amount.

Fortunately, the intercept was found clearly vary in response to deliberate heating of the electrons and so we can use the value of the intercept to extract the temperature. By varying the time when the spilling field is applied, we can measure the temperature as a function of time.

Preliminary measurements show that the plasma does indeed cool rapidly, down to a few K (for an initial 100 K sample) in less than 10 μs. We are in the process of studying the temperature dependence as a function of plasma parameters. Unfortunately this method of temperature measurement becomes unreliable at very low (few K) temperatures, because the self-consistent potential calculation cannot be performed at our current level of sophistication.

6 Open questions and future work

Neutral plasmas at sub-K temperatures are both interesting and challenging, experimentally and theoretically. At the moment we do not have a viable temperature measurement technique in this region. The above spilling technique relies on thermal equilibrium, which we have verified by perturbing the temperature by pumping in energy with an RF field and observing the response. We are aided by the fact that the electron-electron thermalization time decreases with decreasing temperature, so that the electrons can remain in thermal equilibrium as they cool and the density drops. But if the Coulomb coupling constant exceeds 1, everything gets much more complicated. The derivation of the electron equilibration time based on Coulomb collisions becomes invalid, so if and how thermal equilibrium occurs is an open question. The $T^{-9/2}$ three-body recombination scaling also becomes invalid, so it is not even clear whether three-body recombination rates continue to increase as T drops. Yet we continue to observe Rydberg atom formation at densities as low as $10^4 cm^{-3}$ where the n^2 factor in the three-body rate has dropped by a factor of 10^{10}.

Exactly what is happening at long times is clearly one of the outstanding questions. Whether the plasma is becoming strongly-coupled and what the mechanism for atom formation is at long times are unknown. It may be that molecular dynamics simulations can answer some of these questions, but this problem is exceedingly demanding on such techniques due to the wide range of relevant timescales in the computation. To correctly model the electron motion including such things as capture into Rydberg states requires time steps in the fs - ps range. But to model the expansion requires timescales in the 10-100 μs timescales. Coupled with the need for a sufficient number of particles to get a good ensemble average makes the computational demands extreme, beyond current capabilities. It may be possible to use artificial systems, such as much lower mass ions (as was done by the Los Alamos group[3]) to address some of these issues, but it remains a significant computational challenge.

Throughout this we have not addressed what is happening to the ions, except with respect to their expansion velocity. At the initial formation of the plasma, a naive application of the Coulomb coupling constant would suggest that they are very strongly-coupled with $\Gamma \geq 10^3$. But whether this is the case will depend on their interactions with the electrons. The ions will heat on a ms timescale due the thermalization with the electrons, so any effects of the strong coupling are likely to be destroyed. But depending on the parameters, it may be possible that the ions remain strongly coupled into the regime where the electron temperature has gotten so low that it stops imparting much heat to the ions. Perhaps at long times the system can have both strongly-coupled ions and electrons. Experiments at BYU[12] and Rice[13] are being constructed for Ca and Sr respectively, both systems where the ion distribution can be conveniently measured with resonance fluorescence. They may be able to shed some more light onto the behavior of the ions in this system.

Another possibility for the future is to adapt more techniques from plasma physics and try to implement some form of magnetic confinement. Although this would then allow a much longer time to study the plasma, it would also vastly alter the dynamics as adiabatic expansion cooling would be suppressed.

We observed [14] that if we tune the laser to below the ionization limit to excite a cloud of cold Rydberg atoms, it would spontaneously evolve into an ultracold plasma. This was subsequently studied by a number of groups [15, 16, 17] who all came to the conclusion that the process was a sort of avalanche, where a few cold seed electrons would ionize a few Rydberg atoms, creating the conditions for electron trapping and ultracold plasma formation. Once the electrons were trapped, they would have many more opportunities to further ionize Rydberg atoms, leading to the avalanche process. There is still some dispute as to the source of the seed electrons, with blackbody photoionization, Rydberg-Rydberg collisions, and Rydberg-hot atom collisions all being suggested as possibilities. When the plasma is formed from the Rydbergs, its evolution looks indistinguishable to a plasma created a few K above the ionization limit. It will be interesting to measure the temperature of this spontaneously formed plasma, as there is no obvious initial formation energy (in fact it is negative!).

The cold Rydberg gas - plasma transition and the plasma - Rydberg atom formation (in fact a cloud of Rydbergs will evolve into a plasma, which will then produce Rydberg atoms) are signatures of the system residing on the border between single particle atomic physics and the collective behavior of plasma physics. One can ask the question whether there is a difference between a sample of Rydberg atoms being continually formed and reionized and a plasma with strong electron-ion correlations. And what is the effect of expansion and cooling on such a system? There are clearly many unanswered questions and many opportunities for further experimental and theoretical progress in the new field of ultracold plasmas and Rydberg gases. This work was partially supported by the ONR.

References

[1] T. C. Killian *et al.*, Phys. Rev. Lett. **83**, 4776 (1999).

[2] L. Spitzer, Jr. *Physics of Fully Ionized Gases*, Wiley, New York, 1962.

[3] S. Mazevet, L. A. Collins, and J. D. Kress, Phys. Rev. Lett. **88**, 0550001 (2002).

[4] S. G. Kuzmin and T. M. O'Neil, Phys. Rev. Lett. **88**, 065003 (2002).

[5] S. Kulin *et al.*, Phys. Rev. Lett. **85**, 318 (2000).

[6] F. Robicheaux and J. D. Hanson, Phys. Rev. Lett. **88**, 055002 (2002).

[7] T. C. Killian *et al.*, Phys. Rev. Lett. **86**, 3759 (2001).

[8] P. Mansbach and J. Keck, Phys. Rev. **181**, 275 (1969).

[9] The third body does not necessarily have to be an electron in this collision. It could also be an ion or even a non-photoionized neutral atom. Because of the light mass of the electrons and their correspondingly higher average velocity, however, the three-body recombinations with the third body being an electron are expected to dominate.

[10] S. G. Kuzmin and T. M. O'Neil, to appear in Phys. of Plasmas.

[11] Y. Hahn, Phys. Lett. A **293**, 266(2002). Y. Hahn, Phys. Rev. E **64**, 046409 (2001).

[12] S. Bergeson, private communication.

[13] T. Killian, private communication.

[14] S. L. Rolston, S. D. Bergeson, S. Kulin, and C. Orzel, Bull. Am. Phys. Soc.**43**, 1324 (1998).

[15] M. P. Robinson *et al.*, Phys. Rev. Lett. **85**, 4466 (2000).

[16] E. Eyler, Bull. Am. Phys. Soc.**45**, 56 (2000).

[17] S. K. Dutta *et al.* Phys. Rev. Lett. **86**, 3993 (2001).