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Low-frequency electrostatic waves in a magnetized, current-free, heavy negative ion plasma

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Abstract. We report experimental observations of a low-frequency ($\ll$ ion gyrofrequency) electrostatic wave mode in a magnetized cylindrical (Q machine) plasma containing positive ions, very few electrons and a relatively large fraction ($n_-/n_e > 10^3$) of heavy negative ions ($m_-/m_+ \approx 10$), and no magnetic field-aligned current. The waves propagate nearly perpendicular to **B** with a multiharmonic spectrum. The maximum wave amplitude coincided spatially with the region of largest density gradient suggesting that the waves were excited by a drift instability in a nearly electron-free positive ion–negative ion plasma

1. Introduction

Plasmas containing negative ions are found in both naturally occurring plasmas and in plasmas used for technological applications. Negative ions (NO_2^-, O_2^-, O^-) have been detected in the Earth's D and lower E regions at altitudes between 73 and 90 km (Narcisi et al. 1971). Electron 'bite-outs', pronounced density depletions over relatively narrow altitude ranges, observed in the summer polar mesosphere at around 85 km have been associated with capture of electrons by aerosol particles (Reid 1990). Positively charged nanometer-sized particles were observed on a sounding rocket flight in the altitude range from 80 to 90 km in the nighttime polar mesosphere, in a plasma environment having few free electrons and significant numbers of heavy negative ions (Rapp et al. 2005). The positive charge signatures were attributed to charge collection of the more mobile light positive ions, a result predicted by Mamun and Shukla (2003), and observed in the laboratory by Kim and Merlino (2006). Negative ions with masses up to 10 000 amu have also been detected in Titan's ionosphere (Coates et al. 2007). In plasma processing applications, both dc and rf glow discharges in silane are used in the production of silicon-based devices. Multiple negative ion species have been identified in these devices (Howling 1993). The negative ions act as precursors for the formation of large 'dust' particles which, in a processing reactor, can contaminate the product (Choi and Kushner 1993), or can be harvested as ultra-high purity powders (Hahn and Averback 1990).

The presence of negative ions in a plasma can significantly modify dispersion relations of plasma waves, and give rise to new modes (D'Angelo et al. 1966; Wong et al. 1975; D'Angelo and Merlino 1986; Song et al. 1989, 1991; Sato 1994). Many experimental investigations of the effects of negative ions on plasma waves have been performed in Q machines using sulfur hexafluoride. SF_6 (146 amu) has a relatively large cross section for attachment of the low-energy (0.2 eV) electrons in the Q machine, and forms a single, stable parent negative ion SF_6^- (Sheehan and Rynn 1988; Sato 1994; Christophorou and Olthoff 2000). In electropositive plasmas, $m_e/m_+ \ll 1$; however, in negative ion plasmas in which $m_-/m_+ > 1$, $n_-/n_e \gg 1$, and $T_+ > T_-$, it becomes possible to investigate plasma waves in a situation where the positive ions are the more mobile species. Here, m_j, n_j and T_j are the species mass, density and temperature and $j = (e, +, -)$ refer to (electrons, positive ions and negative ions). Heavy negative ion plasmas, with $m_-/m_+ = 350/39$, and $n_e/n_- < 10^{-3}$ were produced in a Q machine by electron attachment to perfluoromethylcyclohexane, C_7F_{14} (Kim and Merlino 2007; Merlino and Kim 2008). Current-driven electrostatic ion-cyclotron (EIC) waves were investigated in this plasma, which contained K^+ positive ions, $C_7F_{14}^-$ negative ions and a small number of free electrons (Kim et al. 2008). The presence of the heavy negative ions lowered the threshold for excitation of multi-harmonic EIC waves to current levels insufficient for wave excitation in the positive ion–electron plasma. The experimental results were in good general agreement with a kinetic instability analysis (Rosenberg and Merlino 2009).

In this paper, we report observations of low-frequency electrostatic waves in a plasma containing substantial amounts of heavy ($m_- \gg m_+$) $C_7F_{14}^-$ negative ions, but with no magnetic field-aligned current. Based on measurements of the plasma and wave properties, the waves are likely due to a drift instability driven by a radial density gradient. A local kinetic theory of drift instability in a positive ion–negative ion plasma, including weak collisional effects, was presented by Rosenberg and Merlino (2013). The effect of negative ions on drift

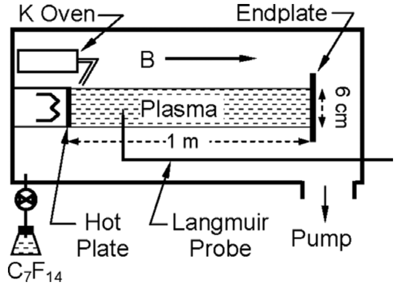

Figure 1. Schematic of the experimental setup. The diameter of the hot plate is 6 cm and the distance from the hot plate to the cold plate is 1 m.

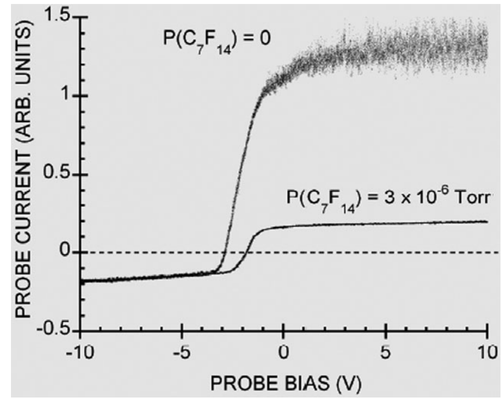

Figure 2. Langmuir probe I–V characteristics before (top trace) and after (bottom trace) C_7F_{14} is added to the plasma.

waves in an Ar/O_2 plasma has also been studied recently by Knist et al. (2011). Their experiments were performed in an RF-produced plasma at neutral pressures ∼0.75–7.5 mTorr and T_e 100 T_+.

In Sec. 2, the details of the experimental device and measurement methods are presented. The results are presented in Sec. 3.1, followed in Sec. 3.2 by a discussion of the possible wave excitation mechanisms. The conclusions are summarized in Sec. 4.

2. Experimental setup and methods

The experiments were performed in the IQ-3 Q machine shown schematically in Fig. 1. The plasma was formed by contact ionization of K atoms from an atomic oven on a 6-cm-diameter tantalum hot plate heated to ∼2300 K. The plasma was confined by a uniform longitudinal magnetic field of 0.1–0.3 T. The background gas pressure in the chamber was $<10^{-6}$ Torr. The plasma column, approximately 3 cm in radius, was terminated on a cold, floating endplate located 1 m from the hot plate. Measurements of the plasma density, electron temperature (equal to the ion temperature) and floating potential were made using a planar Langmuir probe of 4 mm diameter, which could be moved axially and radially in the plasma. The typical plasma parameters were: $T_e \approx T_+ \approx 0.2$ eV, and $n_+ \approx 5 \times 10^{14}$–$2 \times 10^{16}$ m^{-3}. At B = 0.3 T, $T_e = T_+ = 0.2$ eV, the electron and ion gyroradii are $\rho_e = 3.5 \times 10^{-6}$ m and $\rho_+ = 9.5 \times 10^{-4}$ m, respectively.

Negative ions were produced by leaking C_7F_{14} vapor into the vacuum vessel at partial pressures in the range ∼(1×10^{-6}–1×10^{-4}) Torr (Kim and Merlino 2007). The effect of adding C_7F_{14} was observed as a reduction in the electron saturation current to a Langmuir probe in the plasma. Figure 2 shows typical examples of two Langmuir probe I–V curves, the upper one taken with no C_7F_{14}, and the lower one taken with C_7F_{14} at P = 3×10^{-6} Torr. (Note that the positive current shown in these plots is due to the electron or (electron + negative ion) current, while the negative current is the positive ion current.) The effect of electron attachment to C_7F_{14} results in a reduction in the (electron + negative ion) saturation current, while the ion current is unaffected. The current due to the (electrons + negative ions) is due primarily to the free electrons, since their thermal speed is much larger than that of the negative ions. The probe floating potential was more positive when the negative ions were formed.

The type of measurement shown in Fig. 2 is the basis for estimating $\varepsilon \equiv n_-/n_+$, the fraction of negative ions to positive ions in the plasma. Charge neutrality requires $n_+ = n_e + n_-$, so that $\varepsilon = 1 - n_e/n_+$. If the positive ion density remains approximately constant when the negative ions are produced (as in Fig. 2), we can write $n_+ \approx n_{+0} = n_{e0}$, where n_{+0} and n_{e0} are, respectively, the positive ion and electron density in the absence of the negative ions, then $\varepsilon = 1 - n_e/n_{e0}$. The ratio, n_e/n_{e0}, can be determined from the fractional reduction in the saturation current due to negative charges when the negative ions were present, because $I^0_{ns} = I^0_{es} \propto n_{e0}$ and $I_{ns} = I_{es} + I_{nis} \approx I_{es} \propto n_e$, where I_{ns} is the probe saturation current due to (electrons + negative ions), and I_{nis} is the negative ion saturation current which, for $n_-/n_+ < 10^3$ and $T_- \ll T_e$, and $m_-/m_e \gg 1$ is less than I_{es}. Under these conditions, the fraction of negative ions in the plasma can be estimated using $\varepsilon \approx 1 - I_n/I_{n0}$, where I_{n0} is the negative charge carrier saturation current without negative ions (i.e. the electron saturation current) and I_n is the negative charge carrier saturation current with negative ions present. For the data in Fig. 2, $\varepsilon \approx 0.85$; however, as shown in Kim and Merlino (2007) values of $n_e/n_+ < 10^{-4}$ could be obtained using C_7F_{14} at pressures 10^{-5} Torr, so that it was possible to have a nearly electron-free plasma in which the effects of neutral-ion collisions were negligible.

3. Experimental results and discussion

3.1. *Experimental results*

The effect of the negative ions on the radial profiles of the Langmuir probe floating potential is shown in Fig. 3. (The floating potential is a good approximation to the plasma potential in a Q machine.) Figure 3(a) is a typical floating potential profile without the negative ions present. The rapid rise in potential marks the outer edge of the hot plate, with a relatively flat potential over the plasma cross section. When the negative ions

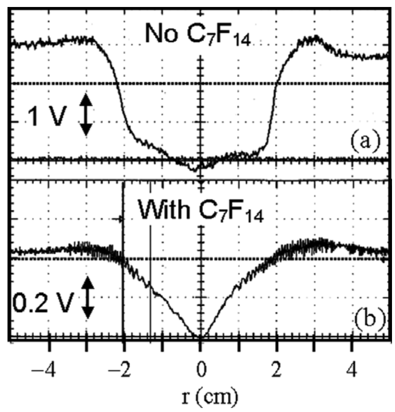

Figure 3. Radial profiles of the probe floating potential (∼plasma potential) before (a: top trace) and after (b: bottom trace) C_7F_{14} is added to the plasma.

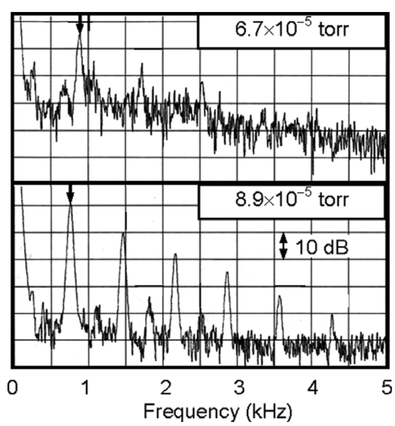

Figure 4. Wave power spectra for $P(C_7F_{14}) = 6.7 \times 10^{-5}$ Torr (upper trace) and 8.9×10^{-5} Torr (lower trace). Arrows mark the fundamental modes.

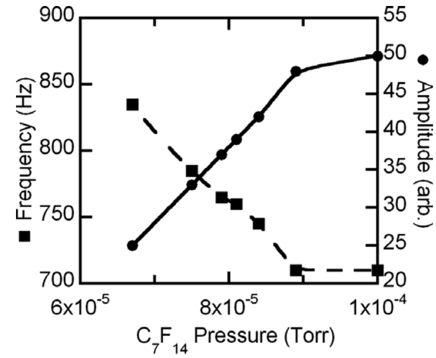

Figure 5. Wave frequency (dashed line) and amplitude (solid line) for various pressures of 6.7×10^{-5}. The relative concentration of negative ions, $n_-/n_{+,}$ increases with $P(C_7F_{14})$.

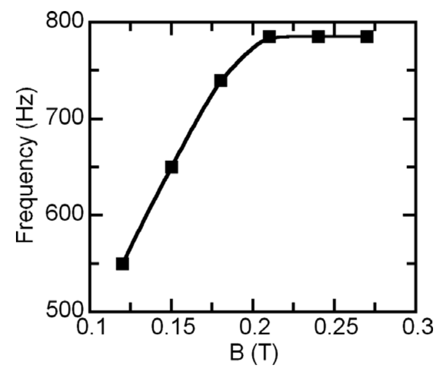

Figure 6. Dependence of the wave frequency for fixed negative ion concentration on magnetic field strength.

were produced (P∼10^{-5} Torr), the typical square wave-type potential profile was transformed into a triangular wave-type profile, with the potential shifting to more positive values in the plasma. The shape of the profile (negative peak in the center) is a possible indication that the negative ions were primarily located near the outside of the plasma column, as observed by Knist et al. (2011).

The spectra of low-frequency electrostatic waves observed in the plasma containing $C_7F_{14}^-$ negative ions are shown in Fig. 4, for C_7F_{14} pressures of (a) 6.7×10^{-5} Torr and (b) 8.9×10^{-5} Torr and for B = 0.18 T. As shown in Fig. 5, as the C_7F_{14} pressure was increased, the wave amplitude increased and higher harmonics were excited, while the frequency decreased. The effect of the magnetic field strength on the wave frequency is shown in Fig. 6. For magnetic fields up to about 0.2 T, the frequency increased, above 0.2 T, the frequency remained approximately constant.

Low-frequency potential fluctuations are also seen in the potential profile in Fig. 3(b). The maximum potential fluctuation level $\tilde{V} \sim 50$ mV corresponded to a wave amplitude, $e\tilde{V}/kT_e \sim 0.25$; this was very close to the value of $\tilde{n}/n_{+0}$ obtained from measurements of the fluctuations, $\tilde{n}$ in the plasma density n_{+0}. The potential fluctuations were largest in the range of 3 cm < r < 2 cm, which corresponds to the region of relatively small radial electric field. The fluctuation amplitude was considerably smaller in the region around the center of the plasma where the largest radial electric fields ∼0.2 V cm^{-1} were present. The maximum wave amplitude corresponded closely to the radial position where $E_r \approx 0$. The radial profile of the ion density is shown in Fig. 7 along with the profile of the wave fluctuation level. These data indicate that the wave amplitude was a maximum in the region of maximum density gradient.

An attempt was made to determine the propagation characteristics of the waves. This was performed by using a narrowband frequency filter to isolate the fundamental mode (∼750 Hz), and observe the relative wave phase difference using two probes located along a diameter of the plasma column on opposite sides of the plasma center. These observations showed that the fluctuations were approximately out of phase (180 degree phase shift) indicating an $m = 1$ azimuthally propagating mode. Due to the limited motional capabilities of the probes it was not possible to measure the phase angle as a function of the azimuthal position. For an $m = 1$ mode, we estimated that the perpendicular wavelength, $\lambda_\theta \approx 2\pi r \approx 2\pi(2-3)\,\text{cm} \approx (13-18)\,\text{cm}$, or, $k_\theta \approx (0.3-0.5)\,\text{cm}^{-1}$. A measurement of the parallel wavelength was also attempted by comparing the wave phase between

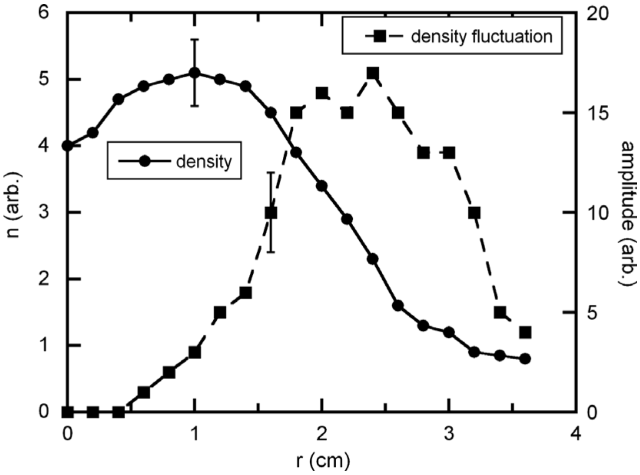

Figure 7. Radial profiles of the plasma density $n(r)$(solid line) and wave amplitude (dashed line).

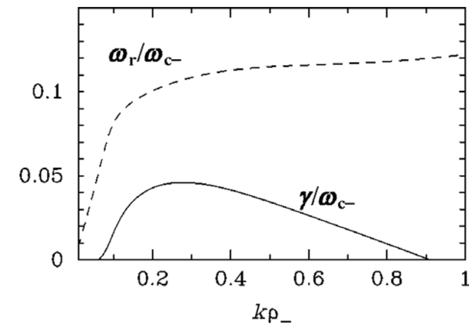

Figure 8. Normalized wave frequency (dashed line) and growth rate (solid line) vs. the normalized wavenumber, $k\rho_-$, based on model by Rosenberg and Merlino (2013).

a probe at a fixed (r^*, z^*) in the plasma column and another probe moving along z but at the same r^*. No clear phase shift could be measured, indicating that the wavelength of the mode along z was most likely much longer than the perpendicular wavelength. If either a half or full wavelength standing wave was excited along z, and the parallel wavelength was limited by the boundary conditions at the ends of the column, then λ_z 1–2 m, or $k_z \approx (0.03 - 0.06)\,\text{cm}^{-1}$. The results then indicate a mode propagating primarily perpendicular to **B.**

3.2. Discussion

In this section, we discuss the likely mechanism responsible for exciting the waves that have been described above. We mention again that there is no magnetic field-aligned current in the plasma that could give rise to wave excitation. In any case, parallel currents would be expected to excite EIC waves or ion acoustic (IA) waves. The observed waves are not EIC waves, since the observed waves frequencies were well below the ion-cyclotron frequencies of either the positive or negative ions. IA waves must be discarded as a possible excitation mechanism, since they would propagate primarily along the magnetic field, and the observed waves propagate nearly perpendicular to **B**. Next, we consider the possibility that the waves could be excited by a Farley–Buneman-type (FB) instability driven by the relative drift between positive and negative ions undergoing **E** × **B** drifts (D'Angelo et al. 1974). This mechanism is also unlikely, since it requires sufficient collisions to produce a relative drift between the positive and negative ions. The neutral pressures in the experiment were so low that the ion-neutral collisions would be ineffective in producing a significant relative drift to excite the FB instability. Furthermore, it was observed that the fluctuations were completely uncorrelated spatially with the largest **E** fields.

The most likely cause of the observed low-frequency fluctuations is a negative ion-modified-drift instability due to the radial density gradient. This conclusion is supported by the observation of the spatial coincidence of maximum wave amplitude and density gradient (Fig. 7), and by the propagation characteristics of the waves.

The drift waves were significantly modified by the presence of the negative ions. In the absence of negative ions, drift waves are typically observed in the 2–10 kHz range. In the presence of the negative ions, the observed drift wave fundamental mode frequency was well below 1 kHz (Fig. 4), and decreased as the concentration of negative ions increased (Fig. 5). For a fixed negative ion concentration, the frequency initially increased as the magnetic field increased, up to a point, and then remained roughly constant (Fig. 6). Both of these properties are in agreement with theoretical predictions based on fluid calculations of drift wave excitation in a multi-ion species plasma (Shukla and Rosenberg 2009). These calculations showed that in the presence of negative ions, a modified drift wave frequency is obtained $\omega_{\text{mod}} = \frac{k_B T_e k_\perp}{eBL_n\left(1+k_\perp^2 \rho_{S,\text{mod}}^2\right)}$, where $\rho_{S,\text{mod}} = \sqrt{\frac{k_B T_e(\varepsilon m_- + m_+)}{e^2 B^2 (1-\varepsilon)}}$ is the effective ion acoustic gyroradius, $k_\perp$ is the perpendicular wave number and L_n is the density gradient scale length. A significant reduction of the drift wave frequency with εoccurs for values of $\varepsilon > 0.99$, or $n_e/n_+ < 10^{-2}$.

The experimental results can also be compared with the linear kinetic theory model of Rosenberg and Merlino (2013) for drift waves in a magnetized, almost electron-free plasma containing positive and negative ions. A plot of the normalized wave frequency and growth rate vs. $k\rho_-$ for $\rho_+/L_n = 0.1$, $m_+/m_- = 39/350$, $T_+/T_- = 7.7$, B $= 0.3$ T and $n_+ = n_-$ 1×10^9 cm^{-3} is shown in Fig. 8. For the experimental parameters $k\ \rho_- = k_\theta\ \rho_- \sim 0.08$, the calculations predict, $\omega_r/\omega_{c-} \approx \sim 0.08$, and with $\omega_{c-} = 2\pi$ (8 kHz) $= 2\pi$ (640 Hz), in reasonable agreement with the observed frequency of 700 Hz. Furthermore, for $k_\theta\ \rho_- \sim 0.08$, the theory predicts modest growth ($\gamma > 0$). It should be noted however, that the experimental parameters are in a regime, where $kL_n \sim 1$, so that a non-local analysis may be more appropriate.

4. Conclusion

An experimental investigation of low-frequency electrostatic waves ($\omega \ll \omega_{c+},\ \omega_{c-}$) in a plasma containing light

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positive and heavy negative ions ($m_- \sim 10m_+$) and a small concentration of electrons has been performed. The plasma was current-free and the wave properties were consistent with a density gradient-driven drift wave instability in a nearly electron-free, positive ion–negative ion plasma. The observations were compared with fluid calculations of drift waves in a plasma with positive ions, negative ions and electrons as well as a kinetic theory of drift waves in an almost electron-free plasma.

Drift waves, which may occur in non-homogeneous plasmas, have been associated with cross-field diffusion in magnetized plasmas. The present experiment may be useful for understanding the behavior of non-homogeneous space and laboratory plasmas containing significant concentrations of heavy negative ions. Finally, we note that experiments in magnetized plasmas containing very heavy negative ions are a step toward studies of dusty plasmas in which the dust particles themselves are also magnetized (Thomas et al. 2012).

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This paper is dedicated to the memory of our dear friend and colleague, Padma Shukla.

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