Angular anisotropy of electron energy distributions in inductively coupled plasmas

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I. INTRODUCTION

Low- and intermediate-pressure inductively coupled plasmas (ICPs) are currently used for etching and deposition in microelectronics fabrication,1–3 fluorescent lighting,4,5 and for the growth of materials such as aligned carbon nanotubes.6 Power in ICPs is largely transferred from the radio-frequency (rf) electric fields to electrons within the electromagnetic skin layer. Many experimental and theoretical studies of this region have shown that the electron energy distributions (EEDs) are generally non-Maxwellian as a result of nonequilibrium transport of electrons.7–10 At the same time, little is known about the angular dependence of the EEDs in and near the skin layer, particularly, when the skin layer is anomalous.11 This is partly a consequence of the difficulty of making electric probe measurements of angle-dependent electron energy distributions (AEEDs).12

The angular anisotropy of the AEE in ICPs is often assumed to be small so that a two-term spherical harmonic expansion can be used in the direct solution of Boltzmann’s equation.8,13,14 This assumption works well for high-pressure or highly collisional plasmas, and for conditions where inelastic collision frequencies are small compared to elastic collision frequencies. These conditions may not be met even in swarm experiments.15 As a result, the two-term approximation becomes increasingly less applicable as the pressure decreases to the regime of interest for microelectronics fabrication (<10 mTorr). For example, measurements of electron drift velocities in the skin layer of an ICP reactor at 10 and 50 mTorr by Meyer et al. were found to be comparable to the local electron thermal velocities, which suggests that the AEEDs were considerably anisotropic.16 Kolobov et al. computationally investigated the angular distribution of electrons in an ICP having a coaxial solenoidal coil.17 They found that the angular distribution of electrons with energies above the plasma potential was anisotropic in the radial–axial (rz) plane and that this anisotropy depends on the radial position (distance from the coil).

In this article, we report on results from a computational investigation of the angular anisotropy of AEEDs in low-pressure ICPs sustained in Ar and Ar/C4F8. Legendre polynomial coefficients describing the angular dependence of kinetically derived AEEDs are derived using sampling techniques in a Monte Carlo simulation. The test reactor is cylindrically symmetric with a flat coil having antenna current oscillating in the azimuthal direction. (θθ refers to the azimuthal–radial plane. rz refers to the radial–axial plane.) We found that there is significant angular anisotropy in the AEEDs in the θθ plane over a wide range of pressures (1–50 mTorr) and frequencies (1.13–13.56 MHz). Angular anisotropy in the rz plane occurs in the skin layer for most conditions and in the bulk plasma only when the skin layer is anomalous. We attributed these two types of anisotropy to the local electron thermal velocities, which suggests that the AEEDs were considerably anisotropic.16 Kolobov et al. computationally investigated the angular distribution of electrons in an ICP having a coaxial solenoidal coil.17 They found that the angular distribution of electrons with energies above the plasma potential was anisotropic in the radial–axial (rz) plane and that this anisotropy depends on the radial position (distance from the coil).

II. DESCRIPTION OF THE MODEL

The model employed in this study is the Hybrid Plasma Equipment Model (HPEM) described in detail in Ref. 10, and references therein. The model consists of three major
modules. The electromagnetics module (EMM) is used to solve Maxwell’s equations for rf magnetic and electric fields. These fields are then used in the electron energy transport module (EETM) where electron transport coefficients and source functions are calculated using the electron Monte Carlo simulation (EMCS). The EMCS is strongly coupled to the EMM as electron currents used in solution of Maxwell’s equations are directly calculated in this module. In large part due to this coupling, the EMCS addresses noncollisional heating, nonlinear electron dynamics, and warm plasma effects, which are significant at low pressure when the skin layer is anomalous.\textsuperscript{9,14,18} Results from the EETM are transferred to the fluid-chemical kinetics module (FKM), which solves the continuity, momentum, and energy equations for densities, momenta, and temperatures of neutrals and charged species, and Poisson’s equation for the electrostatic potential. The sheath at the walls was not explicitly resolved in the solution of Poisson’s equation as the sheath width was considerably smaller than the size of the computational mesh and the skin depth for the pressures and frequencies of interest. These modules are iterated until a converged solution is obtained. Although the EMM and FKM are two-dimensional, the EMCS is fully three-dimensional, and so resolves transport in the \( \theta r \) and \( rz \) planes.

The time-averaged spatially dependent AEEDs are obtained by recording statistics on the energy, location and direction of electron pseudoparticles while their trajectories are advanced in the EMCS. The methods of advancing the pseudoparticle trajectories (employing electron–neutral, electron–ion, and electron–electron collisions), and the manner of recording energies as a function of position, are described in Ref. 10. The angular dependence of the AEEDs obtained from the EMCS are quantified here using a Legendre polynomial expansion. The full anisotropic character of the AEEDs is directly available from the EMCS and could, in principle, be recorded by binning the pseudoparticles in angle as well as energy. Based on past experience in deriving the harmonic time dependence of excitation rates in similar discharges,\textsuperscript{19} we chose the expansion approach as being more robust against statistical noise and more amenable to analysis.

In this method, the AEEDs in the \( \theta r \) or \( rz \) plane, \( f \), are given by

\[
f(\varepsilon, \mathbf{r}, \phi) = \sum_{\ell} a_\ell(\varepsilon, \mathbf{r}) P_\ell(\cos \phi),
\]

where \( \varepsilon \) is the electron energy, \( \mathbf{r} \) is the spatial location, \( P_\ell \) is the \( \ell \)th Legendre polynomial, and \( a_\ell \) is the \( \ell \)th Legendre polynomial coefficient. \( \phi \) is the angle of the electron trajectory with respect to a reference direction, \( \phi_0 \). In the \( rz \) plane, \( \phi_0 \) is aligned with the \( z \) axis pointing up from the substrate to the coils. In the \( \theta r \) plane, \( \phi_0 \) is aligned with the local azimuthal tangent in the direction of the azimuthal electric field. For brevity in the following, \( \mu = \cos(\phi) \).

The raw statistics from which \( a_\ell \) are computed, \( A_\ell(\varepsilon, \mathbf{r}) \), are updated as electron trajectories and are advanced in the EMCS. After each update of the trajectories of the pseudoparticles,

\[
A_\ell(\varepsilon, \mathbf{r}_k) \rightarrow A_\ell(\varepsilon, \mathbf{r}_k) + \sum_j \Delta \Phi_{ikj}
\]

\[
\times \sum_n \left\{ \frac{2n+1}{2} \delta(\mu_n + \frac{1}{2} \delta \mu_j - \mu_j)P_n(\mu_j) \right\},
\]

where

\[
\Delta \Phi_{ikj} = w_j \Delta t_j \delta(\varepsilon_i + \frac{1}{2} \Delta \varepsilon - \varepsilon_j)
\]

\[\times \sum_m \alpha_m \delta(\mathbf{r}_{m+k} + \frac{1}{2} \Delta \mathbf{r}_{m+k}) - \mathbf{r}_j].\]

\( \varepsilon_i \) and \( \mathbf{r}_k \) are the energy and location of the \( i \)th energy bin and \( k \)th spatial mesh cell having widths \( \Delta \varepsilon_i \) and \( \Delta \mathbf{r}_j \). The summations are over \( j \) pseudoparticles and \( n \) bins discretizing angles. The \( \delta \) function isolates the bin where \( \mu_n + \frac{1}{2} \Delta \mu = \mu_j - \frac{1}{2} \Delta \mu_j \). \( \Delta \Phi_{ikj} \) is a coefficient which accounts for binning the particle in energy and position. \( w_j \) is a pseudoparticle-dependent weighting, which accounts for the number of electrons the pseudoparticle represents, and \( \Delta t_j \) is the previous time step. \( \alpha_m \) is a weighting to account for finite-sized-particle distributions. At the end of given iteration through the EMCS, the coefficients, \( a_\ell(\varepsilon, \mathbf{r}_k) \) are obtained from the raw statistics \( A_\ell(\varepsilon, \mathbf{r}_k) \) as

\[
a_\ell(\varepsilon, \mathbf{r}_k) = A_\ell(\varepsilon, \mathbf{r}_k) \int \left[ \sum_i A_0(\varepsilon_i, \mathbf{r}_k) \int_{-1}^{1} P_0 d\mu \right].
\]

III. COLLISIONAL, LINEAR, AND NONLINEAR EFFECTS ON AEEDS IN ICPS

A schematic of the reactor used in this study is shown in Fig. 1 and is patterned after that used by Standaert et al.\textsuperscript{20} The ICP was produced in a cylindrically symmetric chamber (13 cm in radius and 12 cm tall) using a three-turn antenna set atop a quartz window 1 cm thick. Gas was injected through the inlet below the dielectric window and was pumped out from the bottom of the reactor. A metal ring was used to confine plasma. The electron-impact cross sections and heavy particle reaction rate coefficients used in this investigation for Ar are reported in Ref. 10. Those for Ar/C\textsubscript{4}F\textsubscript{8} will be discussed in an upcoming publication.

A. Plasma properties, Legendre coefficients, and AEEDs

An antenna produces a rf magnetic field \( B_{rf} \) in the plane perpendicular to the \( \theta \) axis as shown in Fig. 1. This rf magnetic field creates an inductively coupled azimuthally di-
rected electric field $E_\theta$. There are at least five dominant forces that act upon electrons. The first is the electrodynamic force $-|q|E_\theta$, $q$ is the electron charge, which accelerates electrons in the $\theta r$ plane. The second is the electrostatic force $-|q|E_z$, which accelerates electrons in the $rz$ plane towards the peak of the plasma potential in the middle of discharge. The third is the second-order nonlinear Lorentz force (NLF), $v \times B_{zf}$, produced by the rf magnetic field. $B_{zf}$ is dominantly radially directed under the coils and $v$ has a large azimuthal component due to acceleration by $E_\theta$. The result is that the NLF produces acceleration axially downward for our geometry. Recently, Tasokoro et al. experimentally found evidence for a fourth force $F_{sh}$ resulting from the superposition of the ambipolar electrostatic field and the rf magnetic field $F_{sh} = -E_z \times B_{zf}$. This force is dominantly directed in the $\theta$ direction for our geometry. We found that there is fifth force acting on the electrons in the $\theta r$ plane, which is commensurate with the linear electrodynamic force at low pressures and low frequencies. This force, $F_{sh}^{(3)}$, is due to the rf electric and magnetic fields.

The electron density and temperature $T_e$ are shown in Fig. 2 for the base case conditions (3 mTorr, 400 W, 3.39 MHz) for ICPS sustained in Ar and Ar/C$_4$F$_8 = 70/30$. The peak electron density, which results from the drift of thermal electrons towards the peak of the plasma potential, is $1.2 \times 10^{11}$ cm$^{-3}$ in Ar and $6 \times 10^{10}$ cm$^{-3}$ in Ar/C$_4$F$_8$. The peak plasma potential is 13.4 V in Ar and 13.0 V in Ar/C$_4$F$_8$. The lower electron density in Ar/C$_4$F$_8$ is due in large part to a higher number of electron–electron and electron–heavy particle collisions, $a_1$ and $a_2$ dominate, and $a_3$ is large only for energies above 30 eV. Near the substrate the coefficients are small at energies below the plasma potential, implying isotropic AEEDs. The odd coefficient $a_3$ dominates and $a_3 \approx a_0$ for energies $> 25–30$ eV. Odd Legendre coefficients represent anisotropy in the forward direction, which in this case is aligned downward along the $z$ axis. The large values of the odd coefficients imply that nearly all high-energy electrons are accelerated out of the skin layer into the bulk plasma by the NLF. In the bulk plasma, where the NLF small and electrons experience a large number of electron–electron and electron–heavy particle collision. $a_1$ and $a_2$ dominate, and $a_3$ is large only for energies above 30 eV. Near the substrate the coefficients are small at energies below the plasma potential, implying isotropic AEEDs. $a_2$, which corresponds to the position of the maximum in $E_\theta$, and for three heights. These heights, 11, 8, and 5 cm, are in the electromagnetic skin layer (about 2 cm thick for the base case conditions), bulk plasma, and near the substrate, respectively. (These locations are noted in Fig. 1.) The Legendre coefficients in Ar and Ar/C$_4$F$_8$ show similar trends, which are described as follows.

The expansion coefficients in the $rz$ plane obtained using reference angle $\mu_0$ aligned along the $z$ axis are shown in Fig. 3. In the skin layer, $a_0$ significantly exceeds the other coefficients for energies below the plasma potential, implying that the AEED, involving electrons electrostatically trapped in the plasma, is nearly isotropic. The odd coefficient $a_3$ dominates and $a_3 \approx a_0$ for energies $> 25–30$ eV. Odd Legendre coefficients represent anisotropy in the forward direction, which in this case is aligned downward along the $z$ axis. The large values of the odd coefficients imply that nearly all high-energy electrons are accelerated out of the skin layer into the bulk plasma by the NLF. In the bulk plasma, where the NLF small and electrons experience a large number of electron–electron and electron–heavy particle collision. $a_1$ and $a_2$ dominate, and $a_3$ is large only for energies above 30 eV. Near the substrate the coefficients are small at energies below the plasma potential, implying isotropic AEEDs. $a_2$, which corresponds to the position of the maximum in $E_\theta$, and for three heights. These heights, 11, 8, and 5 cm, are in the electromagnetic skin layer (about 2 cm thick for the base case conditions), bulk plasma, and near the substrate, respectively. (These locations are noted in Fig. 1.) The Legendre coefficients in Ar and Ar/C$_4$F$_8$ show similar trends, which are described as follows.

The first five expansion coefficients for AEEDs for Ar and Ar/C$_4$F$_8$ in the $\theta r$ plane are shown in Fig. 4. The reference angle here is aligned along the $\theta$ axis or the local tangent. The even coefficients $a_i$ dominate at all positions implying that the AEEDs are stretched in both the $+v_z$ and $-v_z$ directions.

The first five expansion coefficients for AEEDs for Ar and Ar/C$_4$F$_8$ in the $\theta r$ plane are shown in Fig. 4. The reference angle here is aligned along the $\theta$ axis or the local tangent. The even coefficients $a_i$ dominate at all positions implying that the AEEDs are stretched in both the $+v_z$ and $-v_z$ directions.
$v_z$ directions, especially for energies above the plasma potential. This azimuthal asymmetry in the AEED is intuitive as one would expect that the harmonic acceleration by $E_\phi$ would produce symmetric anisotropy, that is, even $a_\ell$ dominating. The even coefficients are particularly large in the skin layer where $E_\phi$ peaks. $a_2$ and $a_4$ are proportionally smaller in the bulk plasma and near the substrate due to electron–electron and electron–heavy particle collisions reducing the anisotropy.

The AEEDs in Ar and Ar/C$_4$F$_8$ as functions of the $v_z$ and $v_r$, and $v_\theta$ and $v_r$ velocity components are shown in Fig. 5 in the middle of skin layer for the base case conditions when seven Legendre expansion components are used. The angular distributions of electrons with energies below the plasma potential are nearly isotropic in the $rz$ plane with a small shift in the $v_z$ direction due to the drift of electrons towards the peak of the plasma potential. The anisotropy of the AEEDs in the $v_z$ direction increases with energy in large part due to the NLF, which accelerates high-energy electrons out of the skin layer.

In contrast, the AEEDs obtained in the $\theta r$ plane using the tangent as the reference direction are anisotropic at both low and high energies. At the beginning of a rf cycle, the skin layer is populated by only thermal electrons. As the rf cycle progresses, these low-energy electrons increase in energy and also accumulate anisotropy. The frequency of electron–electron and electron–electron–heavy particle collisions is insufficient at these low pressures to randomize the angular distribution of these electrons. Consequently, the time-averaged AEEDs, which include electrons from different portions of the rf cycle, are anisotropic in all energy ranges.

The angular anisotropy in the low-energy part of AEEDs in the $\theta r$ plane results in proportionally large $\theta$-directed drift velocities in the skin layer. The average drift speed $w_\theta$ can be estimated as $J_{\theta}/en_e$, where $J_{\theta}$ is the amplitude of current density in the $\theta$ direction and $n_e$ is the electron density. For the base case conditions of 3 mTorr $w_\theta \approx 1.3 \times 10^8$ cm/s. The drift velocity decreases with increasing pressure. For example, at 10 mTorr and 200 W, the maximum of drift velocity is $\approx 5.3 \times 10^7$ cm/s. Meyer et al. measured $w_\theta \approx 5 \times 10^7$ cm/s for similar conditions.\textsuperscript{16}
FIG. 5. (Color) AEEDs in the middle of the skin layer, \(r,z\) = (5 cm,11 cm), in the (a) \(r,z\) and (b) \(\theta,r\) planes for the base case conditions for Ar and Ar/C\(_4\)F\(_8\) mixtures. The anisotropy of the AEEDs increases with energy in the \(r,z\) plane in large part due to the NLF. In contrast, the AEEDs in the \(\theta,r\) plane are anisotropic at both low and high energies.

B. Linear and nonlinear forces in ICPs

The angular anisotropy of the AEEDs is attributed to the superposition of linear and nonlinear forces. The linear equation of motion involves the electrodynamic force accounting for electron acceleration in the \(\theta\) direction and collisional damping

\[
F^{(1)}_\theta = m \frac{d\mathbf{v}^{(1)}_\theta}{dt} = -|q| \mathbf{E}_\theta - m \mathbf{v}^{(1)}_\theta \mathbf{v}_m,
\]

where \(\mathbf{E}_\theta = \mathbf{E}_\theta^0 \cos(\omega t)\). This equation is valid for weakly ionized cold plasmas and does not account for the motion of thermal electrons, which is included in the model. \(\mathbf{v}^{(1)}_\theta\), resulting from \(F^{(1)}_\theta\), is then

\[
\mathbf{v}^{(1)}_\theta = -\frac{|q| \mathbf{E}^0_\theta \cos(\omega t - \varphi)}{m (v^2 + \omega^2)^{1/2}},
\]

where \(\varphi = \arctan(\omega/v_m)\). Without collisions the electron’s acceleration due to \(F^{(1)}_\theta\) produces only even coefficients in the \(\theta r\) plane as the electric field alternates between positive and negative values. Collisions randomizing the purely harmonic electron oscillations, yield the phase difference between \(v^{(1)}_\theta\) and \(E_\theta\), and produce odd coefficients. These odd coefficients are rather small compared to the even coefficients at low pressure when electron motion is dominantly collisionless. They are proportionately larger at higher pressures when the collision frequency is larger.

Nonlinear forces, which are weak in the collision-dominated regime, can dominate under collisionless conditions (pressures <10 mTorr).\(^{23}\) The equation of motion using a second-order nonlinear approximation is\(^{23,24}\)

\[
\mathbf{F}^{(2)} = \frac{m}{\omega^2} \frac{d\mathbf{v}^{(2)}_\theta}{dt} = -|q| \mathbf{E}^0_\theta \cos(\omega t - \varphi) - \mathbf{v}^{(1)}_\theta \mathbf{E}_\theta - \mathbf{v}^{(1)}_\theta \mathbf{B}_z \mathbf{e}_z,
\]

where we neglected the collisional damping term and ignored \(B_z\) compared to \(B_{\theta r}\). The second term on the right side of Eq. (6) is typically referred to as the NLF. The change in position \(r^{(1)}_\theta\) can be determined by integrating \(\mathbf{v}^{(1)}_\theta\) with respect to time:

\[
r^{(1)}_\theta(t) = \frac{|q| \mathbf{E}^0_\theta \cos(\omega t - \varphi)}{m \omega (v^2 + \omega^2)^{1/2}}.
\]

Substituting \(r^{(1)}_\theta\) and \(\mathbf{v}^{(1)}_\theta\) in Eq. (6) with their expressions from Eqs. (5) and (7), using that

\[
\frac{\partial \mathbf{E}_\theta(r,z)}{\partial \theta} = 0,
\]

and neglecting the term

\[
(\mathbf{E}^0_\theta \mathbf{v}^0_\theta) \mathbf{E}^0_\theta = -\frac{(E^0_\theta)^2}{r} \mathbf{e}_z,
\]

as it is small everywhere except at the axis, one finds that the first term on the right side of Eq. (1) vanishes and the second term gives a force directed along the \(z\) axis:

\[
\mathbf{F}^{(2)}_z = \frac{-q^2}{m(v^2 + \omega^2)^{1/2}} \mathbf{e}_z \mathbf{E}^0_\theta \mathbf{E}^0_\theta \sin(\omega t) \cos(\omega t - \varphi),
\]

where we used that \(B_z = B_{\theta r} \sin(\omega t)\). In the collisionless case, \(v_m \ll \omega\) and \(\phi = \pi/2\), and Eq. (10) gives

\[
\mathbf{F}^{(2)}_z = \frac{-q^2}{2m\omega} [E^0_\theta B_{\theta r}^0 - E^0_\theta B^0_\theta \cos(2\omega t)].
\]

The velocity \(\mathbf{v}^{(2)}_z\) and change in position \(r^{(2)}_z\) are

\[
\mathbf{v}^{(2)}_z = -\frac{q^2}{2m\omega^2} E^0_\theta B_{\theta r}^0 [\omega t - \sin(2\omega t)/2],
\]

\[
r^{(2)}_z = -\frac{q^2}{4m\omega^2} E^0_\theta B_{\theta r}^0 [\omega^2 t^2 + \cos(2\omega t)/2].
\]

\(\mathbf{F}^{(2)}_z\) consists of time-independent and time-dependent components. The first component accelerates electrons out of the skin layer and produces odd coefficients. The second component oscillating at the second harmonic produces time-
averaged even coefficients. Owing to $F_z^{(2)}$ scaling as $1/\omega$, one should expect that the anisotropy of the AEEDs due to this force would decrease as the frequency increases.

The third-order equation of motion in the collisionless limit is\(^{34}\)

$$F^{(3)} = m \frac{dv^{(3)}}{dt} = - |q| \left( (v_z^{(2)} \nabla) \mathbf{E}_\theta + v_z^{(2)} B_\theta \mathbf{e}_\theta \right). \quad (14)$$

Substituting $r_z^{(2)}$ and $v_z^{(2)}$ in Eq. (14) with their expressions from Eqs. (12) and (13), we find that the third-order force is directed along the tangent and is given by

$$F_\theta^{(3)} = \frac{|q|^3}{4m^2\omega^2} e_\phi E_\theta^0 \left[ B_\theta^0 \right]^2 [\omega^2 t^2 + 2\omega t + \cos(2\omega t)]/2 - \sin(2\omega t), \quad (15)$$

where we used that

$$B_\theta^0 = \frac{1}{\omega} \frac{\partial E_\theta^0}{\partial z}. \quad (16)$$

Due to $F_\theta^{(3)}$ being inversely related to $\omega$, $F_\theta^{(3)}$ is particularly large at low rf frequencies. The ratio of $F_\theta^{(3)}$ to $F_\theta^{(1)}$ in the collisionless regime can be estimated by averaging Eqs. (15) and (4) with $v_\phi = 0$ over half of the rf cycle when the electric field has a single sign. Using the base case plasma conditions and estimating $B_r$ as 3 G at 13.56 MHz and 10 G at 1.13 MHz, one finds that $F_\theta^{(3)}/F_\theta^{(1)} \approx 5$ at 13.56 MHz and $10^3$ at 1.13 MHz. Computational results presented below will support these estimates and show that the third-order nonlinear force can exceed the linear force $F_\theta^{(1)}$. As such, $F_\theta^{(3)}$ can be the dominant force on electrons in ICPs at low frequencies in the collisionless regime and an important mechanism for power deposition.

C. Variations of AEEDs from collisionless to collisional conditions

The just described features of linear and nonlinear forces in ICPs are indicated by the ratios $a_n/a_0$ for different rf frequencies, but otherwise the base case conditions. These results, shown in Fig. 6, are for $r = 5$ cm for heights ranging from the skin layer (11 cm) to near the substrate (5 cm). In the $rz$ plane, the contributions of higher-order terms at 1.13 MHz are larger than those at 13.56 MHz for all positions as $F_z^{(2)}$ increases with decreasing frequency. The odd coefficients are larger than even coefficients in the skin layer at 1.13 MHz and are commensurate with the even coefficients at 13.56 MHz. In the bulk plasma and close to the substrate, the even coefficients at 1.13 MHz are generally larger than the odd coefficients for energies below 25 eV, producing AEEDs elongated in the $+v_z$ and $-v_z$ directions and symmetric with respect to the $r$ axis.

Only even coefficients are shown in the $r\theta$ plane as the odd coefficients are small for both frequencies. The even coefficients are the largest in the skin layer, where $F_\theta^{(1)}$ and $F_\theta^{(3)}$ peak. Here, even coefficients for 13.56 MHz are larger than those at 1.13 MHz for energies below the plasma potential as a consequence of the increased value of the rf electric field and $F_\theta^{(1)}$ in the skin layer. In contrast, at energies above the plasma potential, even coefficients at 1.13 MHz are commensurate with those for 13.56 MHz, implying that the nonlinear forces $F_\theta^{(3)}$ acting on the high-energy electrons exceed the linear force $F_\theta^{(1)}$. In the bulk plasma and close to the substrate, the even coefficients for both frequencies are commensurate at energies below the plasma potential. At higher energies, $a_3/a_0$ is larger for 1.13 MHz, for which the nonlinear force $F_\theta^{(3)}$ is larger.

The ratios $a_n/a_0$ with and without $B_d$ for 1.13 MHz, but otherwise the base case conditions are shown in Fig. 7 for the $rz$ and $r\theta$ planes. Without $B_d$, the NLF in the EMCS and $F_z^{(2)}$ in Eq. (11) are zero and the anisotropy of the AEEDs in the $rz$ plane is due only to the thermal diffusion of electrons towards the peak in the plasma potential. In the middle of the skin layer ($z = 5$ cm), the $a_n/a_0$ in the $rz$ plane with $B_d$ for energies below 20 eV are larger than those obtained without $B_d$. The coefficients are commensurate for energies above 20 eV. These results are a bit counterintuitive. The NLF should accelerate electrons out of the skin layer, and so odd coefficients with $B_d$ should dominate, which is what we observe at low energy. The large odd $a_n/a_0$ at higher energies without $B_d$ are likely a consequence of being close to the...
boundary of the plasma. High-energy electrons (above the plasma potential), moving vertically upwards, which would contribute to even \( a_n/a_0 \), are lost from the plasma leaving only those directed downward to contribute to odd \( a_n/a_0 \). In the middle of the plasma and near the substrate (\( z=8 \) and 5 cm) the lack of significant \( a_n/a_0 \) at low energies implies that without \( B_{rf} \) the AEEDs are fairly isotropic. Any directionality at these energies is due to residual effects of the NLF, which accelerate electrons out of the skin layer.

The anisotropy in the \( \theta r \) plane is determined by the superposition of the linear \( F^{(1)}_{\theta} \) and nonlinear force \( F^{(3)}_{\theta} \) given by Eqs. (4) and (15), respectively. \( F^{(1)}_{\theta} \) is not affected by \( B_{rf} \), whereas \( F^{(3)}_{\theta} \) is directly proportional to \( B_{rf} \). Consequently, \( a_n/a_0 \) with \( B_{rf} \) are similar to those without \( B_{rf} \) for energies below the plasma potential as electrons with these energies are electrostatically trapped in the plasma, and the anisotropy of their distribution is determined by \( F^{(1)}_{\theta} \). Above the plasma potential, the coefficients with \( B_{rf} \) are significantly larger than those without \( B_{rf} \), implying that the anisotropy of AEEDs at high energies is determined by \( F^{(3)}_{\theta} \).

The differences in Legendre coefficients with and without \( B_{rf} \) could be in part due to the force \( \mathbf{F}_{sh} \sim \mathbf{E}_s \times \mathbf{B}_{rf} \) reported by Tasokoro et al., and which is due to the electrostatic ambipolar field. For our conditions, one would expect that \( \mathbf{F}_{sh} \) is only significant in the presheath (about 1 cm from walls for the base case) where the gradient of the plasma potential and \( B_{rf} \) are largest. \( \mathbf{F}_{sh} \) likely affects, on a fractional basis, low-energy electrons most severely as the high-energy electrons quickly transverse through the presheath. Since inclusion of \( B_{rf} \) affects the Legendre coefficients at high energies most severely, as shown in Fig. 7, one might conclude that the angular anisotropy of AEEDs is not particularly sensitive to \( \mathbf{F}_{sh} \).

Noncollisional heating, nonlinear electron dynamics, and warm plasma effects are significant at low pressure, when the skin layer is anomalous, and are weak at high pressure, when the collision frequency is large. Consequently, the Legendre coefficients change significantly with pressure, as shown in Fig. 8. At 1 mTorr, when the NLF and warm plasma effects are important, \( a_3 \) in the \( rz \) plane is large at high energies, implying there is anisotropy in the AEEDs in the \( -v_z \) direction. The coefficients at 50 mTorr, when the collision frequency is large, are an order of magnitude smaller than \( a_0 \), implying a more isotropic distribution. Note that the coefficients slowly decrease with energy at 1 mTorr, whereas those at 50 mTorr exponentially decrease as their energy increases. These trends can be explained by the different mechanisms for electron heating at 1 and 50 mTorr. At 50 mTorr, the electrons are highly collisional and Ohmic
heating dominates, which yields electron distributions with a low-energy tail. At 1 mTorr, collisionless heating dominates over Ohmic heating and forms EEDs with a high-energy tail.25,26

The even coefficients in the $\theta r$ plane are large at both 1 and 50 mTorr, implying that there is anisotropy in the AEEDs over a wide range of pressures originating from both $F_\theta^{(1)}$ and $F_\theta^{(3)}$. The even coefficients at 1 mTorr are larger than those at 50 mTorr, as the nonlinear force $F_\theta^{(3)}$, acting on the high-energy electrons, is larger at low pressures and weaker at high pressures. In contrast, the odd coefficients increase with pressure as they originated from the collisions, which randomize the purely harmonic electron oscillations and produce the phase difference between $v_\theta$ and $E_\theta$.

IV. CONCLUDING REMARKS

The anisotropy of AEEDs in low-pressure ICPs was investigated using Monte Carlo techniques by sampling the trajectories of the electrons and computing Legendre coefficients. The AEED is anisotropic in the $rz$ plane, favoring the high-order odd coefficients. The $-v_z$ component dominates at low frequencies and pressures due largely to the nonlinear Lorentz forces. The anisotropy is largest at higher energies. In the $\theta r$ plane, even coefficients dominate, implying a large $w_\theta$ drift velocity in the azimuthal electric field. We found that the anisotropy in the $\theta r$ plane is due to electron acceleration by linear electrodynamic and nonlinear third-order forces. Anisotropy in the $rz$ plane dominantly occurs when the skin layer is anomalous, whereas anisotropy in the $\theta r$ plane persists to higher pressures. For operating conditions typical of plasma processing reactors, higher Legendre coefficients in both the $rz$ and $\theta r$ planes have significant values.

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