Instability-Enhanced Collisional Effects and Langmuir's Paradox

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Anomalously fast equilibration of the electron distribution function to a Maxwellian in gas-discharge plasmas with low temperature and pressure, i.e., Langmuir's paradox, may be explained by electron scattering via an instability-enhanced collective response and hence fluctuations arising from convective ion-acoustic instabilities near the discharge boundaries.

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The scattering of electrons in low-pressure gas discharges has presented an anomaly since the earliest days of plasma physics research. A seminal paper by Langmuir [1], reported measurements showing that the electron velocity distribution function (EVDF) equilibrated to a Maxwellian much closer to the discharge boundaries (which selectively remove high energy electrons) than could be explained by the kinetic theory of scattering via their individual Coulomb electric fields. In this Letter, we consider details of the plasma-boundary transition and show that an instability-enhanced collective response and hence fluctuations, due to ion-acoustic instabilities in the presheath of these discharges, causes electron-electron scattering to occur much more frequently than it does by Coulomb interactions alone.

This collective response arises from the instability amplification of thermal fluctuations; it effectively extends the range over which particles interact beyond the Debye sphere in which Coulomb interactions are confined. Since these ion-acoustic instabilities convect out of the plasma before reaching nonlinear levels, turbulence theories are not applicable in describing the enhanced electron scattering. The Boltzmann $\mathcal{H}$ theorem remains valid when this collective response is present [2] and a Maxwellian is the unique equilibrium which is established at a rate rapid enough to be consistent with Langmuir's measurements.

In his work developing the fluorescent lamp, Langmuir often used a spherical discharge tube approximately 3 cm in diameter which was made of glass and energized by electrons emitted from a hot filament [1]. He discovered that nearly all of the discharge was a quasineutral plasma but that because electrons diffused to the boundaries much faster than the more massive positively charged ions, a thin electric field, which he named a sheath, surrounded the plasma and acted to reflect most of the incident electrons so that the electron and ion fluxes balanced at the boundary. For such an ambipolar ion sheath, the potential drops $e\Delta\phi_s \approx -T_e \ln\sqrt{2\pi m_e/M_i}$ ($\approx 5T_e$ for mercury) in only a few electron Debye lengths $\lambda_{\rm De} \equiv \sqrt{T_e/4\pi e n_e}$. It was later shown that an additional, but much weaker, presheath electric field is also present which accelerates the ion fluid speed to the sound speed, $V_i \geq c_s \equiv \sqrt{T_e/M_i}$ at the presheath-sheath boundary. This result is commonly attributed to Bohm [3], but it was also appreciated in Langmuir's earlier works deriving the "plasma balance equation" [4]. The potential in the presheath of these discharges typically drops $e\Delta\phi_{ps} \approx T_e/2$ over a distance characteristic of the ion-neutral collision mean free path $\lambda^{i/n} \gg \lambda_{\rm De}$.

In his apparatus, Langmuir measured the EVDF using an electrostatic probe (now called a Langmuir probe) and found that it was Maxwellian at all velocities despite the fact that his calculated electron-electron scattering collision length $\lambda^{e/e}$ was much longer than the tube diameter. Langmuir expected significant depletion of the EVDF for $v_\| \gtrsim v_{\|c} \equiv \sqrt{2e\Delta\phi_s/m_e}$. Here the $\|$ direction is parallel to the sheath electric field (perpendicular to the bounding surface). It was unexplainable how his discharge could remain lit because the vast majority of ionization events were attributed to the very same electrons in the tail of the Maxwellian EVDF (rather than filament emitted electrons) that his theory predicted to be missing. Langmuir's measurements implied that some anomalous mechanism for electron scattering must have been present which was capable of boosting the velocity of many electrons and rapidly establishing the Maxwellian equilibrium. His measurements and their implication were later named Langmuir's paradox by Gabor *et al* [5] and today remain a serious discrepancy in the kinetic theory of gas discharges. Analogies to it have also since been drawn in the context of the evolution of galaxies as gravitational plasmas [6].

Several ideas have been proposed attempting to explain Langmuir's paradox. These include the possibility of circuit oscillations interfering with probe measurements of the EVDF [1], scattering of electrons by photons [1] and by sheath oscillations [5], and due to the electronic polarizability of neutral gas atoms or molecules [7]. More recent work [8] has developed a "nonlocal approximation" to electron kinetics claiming that there may be no paradox. None of the previous work has given a definitive answer to Langmuir's paradox.

Oscillations in the MHz frequency range have been measured near sheaths [5], but no theory has been proposed to describe their origin or suggest how they lead to quick equilibration to a Maxwellian. We propose that these oscillations are ion-acoustic instabilities, which are in the MHz range. To show that they lead to a Maxwellian we employ a theory derived in Ref. [2] that extends the Lenard-Balescu kinetic theory [9] to include instability-enhanced fluctuations that have not reached a nonlinear level. Thus the theory is applicable for absolute instabilities in a finite temporal domain, or for convective instabilities in a finite spatial domain. This theory is well suited to describing the effects of ion-acoustic instabilities in the presheath because they convect out of the plasma while still in their linear growth phase.

Other instabilities may also be present and lead to instability-enhanced collisional effects. For example, filamentary discharges, such as Langmuir's, have a small population of high energy electrons that may cause a bump-on-tail instability. Two-stream instabilities may be present in presheaths when there are multiple species of ions, or if some ions are multiply charged. However, ion-acoustic instabilities are universal in presheaths of gas discharges with low temperature and pressure.

We apply the kinetic theory to the same discharge parameters that Langmuir used in his original experiments [1]. This was a mercury plasma with electron (plasma) density $n_e \approx 10^{11}$ cm^{-3}, neutral density $\approx 10^{13}$ cm^{-3} (0.3 m Torr), and ion and electron temperatures of $T_i \approx$ 0.03 eV and $T_e \approx 2$ eV, respectively.

The evolution of the distribution function for any species s is governed by the plasma kinetic equation $df_s/dt = \sum_{s'} C(f_s, f_{s'})$, in which $d/dt = \partial/\partial t + \mathbf{v} \cdot \partial/\partial \mathbf{x} + \mathbf{E} \cdot \partial/\partial \mathbf{v}$ is the convective derivative. Here, $C(f_s, f_{s'})$ is a collision operator describing the evolution of f_s due to collisions with each plasma species s' including itself ($s = s'$). The collision frequency thus scales with the magnitude of the collision operator $\nu_{s/s'} \sim C(f_s, f_{s'})/f_s$.

The Lenard-Balescu equation [9] provides an accurate collision operator when the plasma is stable; it captures the physics of Coulomb interactions between individual particles as well as effects that arise from the collective response described by a general plasma dielectric function. However, when applied to this discharge, it predicts a scattering collision length similar to Langmuir's estimate and thus cannot explain the paradox. The conventional results obtained from Fokker-Planck [10] or Landau theory [11] can be obtained from Lenard-Balescu theory assuming an adiabatic dielectric response, $\hat{\varepsilon} \approx 1 + 1/k^2\lambda_{\mathrm{De}}^2$.

In Ref. [2] the Lenard-Balescu formalism was extended to allow for unstable plasmas as long as the corresponding fluctuation levels of the density and electric field remain low enough that a linear perturbation analysis is valid. These effects were included in the derivation by allowing positive growth rates which result when the imaginary part of the roots of the dielectric function are positive $\hat{\varepsilon}(\mathbf{k}, \omega_j) = 0 \Rightarrow \mathrm{Im}\{\omega_j(\mathbf{k})\} > 0$. After inverting Laplace transforms, terms that scale as $\exp(-i\omega_j t)$ emerge and produce an instability-enhanced collective response.

The resulting collision operator is

$$C(f_s, f_{s'}) = -\frac{\partial}{\partial \mathbf{v}} \cdot \int d^3v' \mathbf{Q}(\mathbf{v}, \mathbf{v}') \cdot \left(\frac{1}{m_{s'}}\frac{\partial}{\partial \mathbf{v}'} - \frac{1}{m_s}\frac{\partial}{\partial \mathbf{v}}\right) \times f_s(\mathbf{v}) f_{s'}(\mathbf{v}') \quad (1)$$

in which

$$\mathbf{Q}(\mathbf{v}, \mathbf{v}') = \frac{2q_s^2 q_{s'}^2}{m_s} \int d^3k \frac{\mathbf{k}\mathbf{k}}{k^4} \delta(\mathbf{k} \cdot (\mathbf{v} - \mathbf{v}')) \times \left[\frac{1}{|\hat{\varepsilon}(\mathbf{k}, \mathbf{k} \cdot \mathbf{v})|^2} + \sum_j \frac{\pi \delta(\omega_{R,j} - \mathbf{k} \cdot \mathbf{v}) e^{2\gamma_j t}}{\gamma_j |\partial \hat{\varepsilon}(\mathbf{k}, \omega)/\partial \omega|^2_{\omega_{R,j}}}\right] \quad (2)$$

is the collisional kernel. The first term in Eq. (2) constitutes the Lenard-Balescu equation where collective interactions in a stable plasma are described by $|\hat{\varepsilon}(\mathbf{k}, \mathbf{k} \cdot \mathbf{v})|^2$. In a low-pressure gas-discharge plasma, electrons are typically adiabatic so the Lenard-Balescu equation reduces to the Fokker-Planck or Landau's equation. The second term in Eq. (2) describes the enhanced collective response that arises when instabilities amplify the background thermal fluctuations of the plasma. This term grows exponentially according to the growth rate γ_j of each excited mode j with real frequency $\omega_{R,j}$. Before the instability amplification, the second term is typically $\sim 1/\ln\Lambda$ ($\Lambda = \lambda_{\mathrm{De}}/b_{\min} \approx 12\pi n_e \lambda_{\mathrm{De}}^3$) smaller than the first term; but as the instabilities grow, it can eventually dominate.

According to Eq. (2), the collisional kernel and the collision operator can be described as the sum of contributions from stable plasma interactions and instability enhancements. Thus we write $\mathbf{Q} = \mathbf{Q}_{\mathrm{LB}} + \mathbf{Q}_{\mathrm{IE}}$ where the subscript LB refers to the Lenard-Balescu contribution and IE to the instability-enhanced contributions to the collisional kernel.

The collision operator of Eqs. (1) and (2) obeys [2] many of the same physical properties as the Lenard-Balescu collision operator [9]. These include conservation of density, momentum, and energy as well as Galilean invariance and the Boltzmann $\mathcal{H}$ theorem. The Boltzmann $\mathcal{H}$ theorem states that $d\mathcal{H}/dt \leq 0$, in which $\mathcal{H} \equiv \int d^3v f(\mathbf{v}) \times \ln f(\mathbf{v})$ is the $\mathcal{H}$ functional (entropy increases until equilibrium). Furthermore, and most importantly here, the Boltzmann $\mathcal{H}$ theorem was extended [2] to show that a Maxwellian is the unique equilibrium solution. Thus, the plasma evolves to a Maxwellian not only in stable plasmas, but also in plasmas with linear instabilities.

Because it is the equilibrium solution, the collision operator for electron-electron collisions vanishes [$C(f_e, f_e) = 0$] if f_e is a Maxwellian. Estimating $\partial/\partial \mathbf{v} \sim 1/v_{Te}$, where $v_{Te} \equiv \sqrt{2T_e/m_e}$ is the electron thermal speed, Eq. (1) shows that the electron-electron collision frequency scales as

$$\nu^{e/e} \sim \frac{n}{m_e v_{Te}^2}(Q_{\rm LB}^{e/e} + Q_{\rm IE}^{e/e}) \tag{3}$$

in which the scalars $Q_{\rm LB}^{e/e}$ and $Q_{\rm IE}^{e/e}$ represent the dominant contributions of the dyads $\mathbf{Q}_{\rm LB}^{e/e}$ and $\mathbf{Q}_{\rm IE}^{e/e}$. Evaluating Eq. (2) for the Lenard-Balescu term using an adiabatic electron response yields

$$\mathbf{Q}_{\rm LB}^{e/e} \approx \frac{2\pi e^4 \ln\Lambda}{m_e}\left(\frac{u^2 I - \mathbf{uu}}{u^3}\right) \tag{4}$$

in which $\mathbf{u} \equiv \mathbf{v} - \mathbf{v}'$ and the short wavelength limit of the k-space integral was cut off at $1/b_{\min}$ where $b_{\min}$ is the minimum impact parameter [10]. Since $u \sim v_{Te}$, the collision frequency due to particle-particle interactions is

$$\nu_{\rm LB}^{e/e} \sim \frac{\omega_{pe}}{8\pi n \lambda_{\rm De}^3} \ln\Lambda. \tag{5}$$

For Langmuir's characteristic plasma $\lambda_{\rm LB}^{e/e} \approx v_{Te}/\nu_{\rm LB}^{e/e} \approx$ 28 cm.

A more rigorous analysis reveals that $\nu_{\rm LB}^{e/e} \sim 1/v^3$, as does $\nu_{\rm IE}^{e/e}$. Although we are interested in the truncated tail which concerns electrons with a speed a few times that of v_{Te}, for simplicity we calculate $\nu^{e/e}$ at $v \approx v_{Te}$. Since both the Coulomb and instability-enhanced contributions have the same $1/v^3$ speed dependence, this estimate can be used to compare the relative contribution from each scattering mechanism.

Estimating $Q_{\rm IE}^{e/e}$ requires the plasma dielectric, which for low-pressure gas discharges is

$$\hat{\varepsilon} = 1 + \frac{1}{k^2\lambda_{\rm De}^2} - \frac{\omega_{pi}^2}{(\omega - \mathbf{k}\cdot\mathbf{V}_i)^2} + i\frac{\sqrt{\pi}}{k^2\lambda_{\rm De}^2}\frac{\omega}{k v_{Te}}, \tag{6}$$

in which we assume that the wave phase speed is slow compared to the electron thermal speed, $\omega/kv_{Te} \ll 1$, and fast compared to the ion thermal speed, $(\omega - \mathbf{k}\cdot\mathbf{V}_i)/kv_{Ti} \gg 1$. We have also used the essential property of the example plasma that $T_i/T_e \ll 1$, which implies negligible ion Landau damping.

Estimating $Q_{\rm IE}^{e/e}$ also requires the dispersion relation, defined by the roots of the plasma dielectric, which in this case represent one growing and one damped ion-acoustic mode

$$\omega_{\pm} = \left(\mathbf{k}\cdot\mathbf{V}_i \pm \frac{kc_s}{\sqrt{1 + k^2\lambda_{\rm De}^2}}\right)\left(1 \mp i\frac{\sqrt{\pi m_e/8M_i}}{(1 + k^2\lambda_{\rm De}^2)^{3/2}}\right). \tag{7}$$

We will use the notation $\omega = \omega_R + i\gamma$ where a positive γ represents a growth rate. A growing solution thus exists as long as the ion fluid speed is large enough: $|\mathbf{k}\cdot\mathbf{V}_i| > kc_s/\sqrt{1 + k^2\lambda_{\rm De}^2}$. In deriving Eqs. (6) and (7), we have assumed a Maxwellian EVDF. A solution accounting for direct electron loss for $v_\| \geq v_{\|c}$ was also calculated, but resulted in only negligibly small $O(\exp(-v_{\|c}^2/v_{Te}^2) \times v_{Te}/v_{\|c}) \ll 1$ corrections.

As described in detail in Ref. [2], the $\exp(2\gamma t)$ term in Eq. (2) must be calculated in the rest frame of the unstable mode. Since the ion-acoustic instability is convective,

$$2\gamma t = 2\int_{\mathbf{x}_o(\mathbf{k})}^{\mathbf{x}} d\mathbf{x}' \cdot \frac{\mathbf{v}_g \gamma}{|\mathbf{v}_g|^2}, \tag{8}$$

in which $\mathbf{v}_g \equiv \partial\omega_R/\partial\mathbf{k}$ is the group velocity, $\mathbf{x}_o(\mathbf{k})$ is the location in space where the mode wave vector $\mathbf{k}$ becomes unstable, and the integral $d\mathbf{x}'$ is taken along the path of the mode. An important consequence is that, since ω_- and $\mathbf{x}_o$ have no explicit time dependence, f_e will change with position, but not in time, in the laboratory frame. The plasma can thus remain in a steady state and the EVDF will equilibrate to a Maxwellian at a distance from the sheath determined by $\lambda^{e/e}(\mathbf{x})$.

In principle, the spatial integral in Eq. (8) requires integrating the profile of γ and $\mathbf{v}_g$, which change through the presheath due to variations in the ion fluid speed and the electron density, as well as knowing the spatial location $\mathbf{x}_o(\mathbf{k})$ at which each wave vector $\mathbf{k}$ becomes excited. In estimating Eq. (8) we assume that changes due to spatial variations are weak, and we account for $\mathbf{x}_o(\mathbf{k})$ by only integrating over the unstable $\mathbf{k}$ for each spatial location $\mathbf{x}$. Following these approximations we obtain $2\gamma t \approx 2x\gamma/v_g$.

Using Eq. (3) and this estimate in the k integral in Eq. (2) leads to an effective collision frequency due to instability-enhanced collective interactions:

$$\nu_{\rm IE}^{e/e} \sim \frac{\nu_{\rm LB}^{e/e}}{8\ln\Lambda}\frac{1 + 2\kappa_c^2}{(1 + \kappa_c^2)^2}\exp\left(\eta\frac{x}{l}\right), \tag{9}$$

in which $\kappa_c \equiv \sqrt{c_s^2/V_i^2 - 1}$ accounts for the k-space cutoff and is valid for $V_i \leq c_s$; otherwise $\kappa_c = 0$, and $\eta \equiv l\sqrt{\pi m_e/16M_i}/\lambda_{\rm De}$. Here, l is a length scale characterizing the presheath; typically it is the ion-neutral collision mean free path. The corresponding electron-electron collision length is $\lambda_{\rm IE}^{e/e} \approx v_{Te}/\nu_{\rm IE}^{e/e}$. The location $x = 0$ corresponds to the spatial location where instability onset occurs.

Finally, we apply our estimates of the collision frequency to the presheath using a 1D modified mobility-limited flow model due to Riemann [12]

$$\frac{e\phi}{T_e} = \ln\left(\frac{c_s}{V_i}\right) \quad \text{and} \quad dx = dV_i\left(\frac{c_s^2 - V_i^2}{V_i^2 \nu^{i/n}}\right) \tag{10}$$

to solve for the ion fluid flow profile which determines $\kappa_c(x)$. This model follows from the two fluid equations assuming Boltzmann electrons and quasineutrality. It has been verified experimentally to a distance $x \approx 2l$ [13],

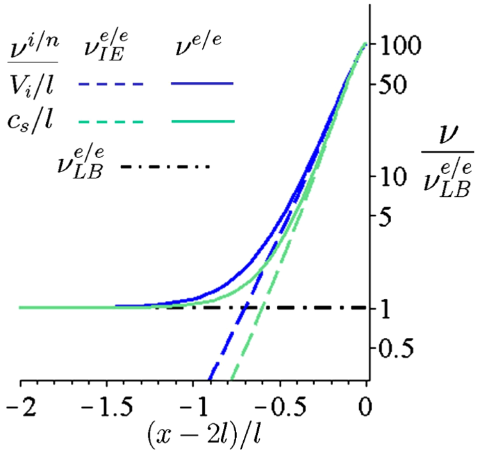

FIG. 1 (color online). Total electron-electron collision frequency normalized to $\nu_{\rm LB}^{e/e}$ (solid lines) with contributions from Coulomb interactions in a stable plasma (black, dash-dotted line) and from instability-enhanced collective interactions (dashed lines). Two common presheath models are represented: $\nu^{i/n} = V_i/l$ (blue) and $\nu^{i/n} = c_s/l$ (aquamarine).

beyond which the ion fluid speed is so slow that any additional contribution to $\nu_{\rm IE}^{e/e}$ is negligibly small. Two cases of Eq. (10) are commonly considered: constant mean free path for ion-neutral collisions $\lambda^{i/n} = l$, $\nu^{i/n} = V_i/l$, or a constant collision frequency $\nu^{i/n} \approx c_s/l$. For these discharge parameters $l \approx 11$ cm [13], and $\eta \approx 4.5$. For each of these, we plot in Fig. 1 the total predicted electron-electron scattering collision length along with the individual contributions from stable plasma theory and the instability enhancement. The theory is not sensitive to which presheath model is used.

It was shown in Ref. [2] that the limits of validity for the scattering theory depend on the number of particles in a Debye cube, $n\lambda_{\rm De}^3$, and $\eta x/l$. For this plasma with $n\lambda_{\rm De}^3 \approx 3 \times 10^3$, the theory is valid for $\eta x/l \lesssim 55$. In this presheath example the maximum $\eta x/l \lesssim 10$, which is reached at the sheath-presheath boundary; thus, the theory is well suited to this problem.

Figure 1 shows that near the sheath-presheath boundary ion-acoustic instabilities enhance the electron-electron scattering approximately 100 times the nominal stable plasma rate. The collision length for electron-electron scattering is shortened by more than a factor of 10 over a distance of approximately $l/2$. Thus, near the plasma boundary, instability-enhanced collective interactions determine the scattering rate and drive the plasma toward the unique Maxwellian EVDF within the presheath length scale, which is consistent with Langmuir's measurements.

Aspects of the model proposed in this Letter can be directly tested experimentally. The k-space fluctuations could be characterized in the presheath. We predict that modes satisfying $k \gtrsim 1/\lambda_{\rm De}$ become unstable and grow exponentially toward the boundary. These fluctuations should disappear due to ion Landau damping if the ions are heated to $T_i \approx T_e$. Alternatively, accounting for ion-neutral damping results in a $-i\nu^{i/n}/2$ term to be added to Eq. (7). Using $\nu^{i/n} \approx c_s/\lambda^{i/n}$, leads to the result that the ion-acoustic instabilities are ion-neutral damped for $\eta \lesssim 1$. Since $\eta > 1$ is required for instability-enhanced scattering, this also represents a maximum neutral density above which the presheath length is so short that the instabilities have an insufficient distance to grow before reaching the boundary. Experimentally, electron scattering could thus be attributed to instability-enhanced collective interactions by measuring both the fluctuations and the EVDF with and without instabilities.

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