Nonequilibrium Phenomena in Thermal Plasmas

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Abstract: Thermal plasmas are used in diverse applications such as spraying, cutting, welding, metallurgy, chemical synthesis, and resource recovery. Thermal plasma flows involve interactions with working gas streams, gas environment, confining device, and processing materials. These interactions lead to the establishment of kinetic (microscopic) and dissipative (macroscopic) nonequilibrium phenomena. Such phenomena can be desirable, e.g. when it leads to greater process efficiency; or detrimental, e.g. when it limits process uniformity. This talk will present an overview of nonequilibrium phenomena in thermal plasmas, with particularly emphasis on their computational description.

Keywords: Dissipative structures, particle kinetics, non-LTE, instability, pattern formation.

1. Introduction

Thermal plasmas are at the core of a wide variety of industrial applications, such as spraying, cutting, welding, metallurgy, nanoparticle production, chemical synthesis, waste treatment, resource recovery, among others. Those applications exploit some of the unique characteristics of thermal plasmas, including high temperatures (~10^4 K), electron densities (~10^{23} m^{-3}), and heat fluxes (~10^5 kW-m^{-2}) [1], which allow high process throughout and the processing of high melting point materials. These characteristics necessarily imply the occurrence of large property gradients wherever the thermal plasma interacts with its surroundings, such as a processing gas stream, gas environment, electrodes, confining devices, and processing materials. These interactions typically lead to the establishment of a state of nonequilibrium in the system. Nonequilibrium phenomena in thermal plasma flows, as in other multiphysics and multiscale systems, can be characterized as *kinetic* or *dissipative*.

Kinetic nonequilibrium is the result of microscopic imbalances, and dissipative nonequilibrium the result of macroscopic imbalances. The computational simulation of nonequilibrium kinetic and dissipative phenomena can provide important information to complement experimental observations, and guide equipment design and process optimization. Nevertheless, the computational description of nonequilibrium phenomena faces diverse, often compound, challenges. These challenges are generally tackled by computational methods (numerical discretization and solution approaches) aimed for multiphysics and multiscale problems and can be broadly categorized in terms of model *fidelity* and numerical *accuracy* [2]. Fidelity refers to the degree of underlying phenomena captured by the model, and therefore is directly related to the number of independent variables in the model. Accuracy refers to the precision of the numerical solution of the model equations, and hence is directly related to the number of discrete unknowns in the problem. In general, fidelity is a major challenge in multiphysics problems, whereas accuracy is a major challenge in multiscale problems. An important characteristic of thermal plasma flows is that their comprehensive computational simulation often stresses the need for both, fidelity and accuracy. The occurrence of kinetic and dissipative nonequilibrium in a thermal plasma system, i.e. the free-burning arc, is schematically depicted in Fig. 1.

2. Kinetic Nonequilibrium

Kinetic nonequilibrium is the result of microscopic particle and field imbalances, as described by particle kinetics frameworks. These imbalances lead to deviations in particle population and energy distribution functions, mass action laws, quasi-neutrality, etc.. A representative manifestation of kinetic nonequilibrium is observed in the region near the anode of a thermal plasma, often known as the *anode sheath*, depicted in the left portion of Fig. 1.

A key characteristic of thermal plasmas is that their constituent particles (electrons, ions, atoms, molecules) are close to a state of Local Thermodynamic Equilibrium (LTE). Nevertheless, it is now well-established that LTE is an adequate approximation within the core of the plasma only, and that significant deviations are found in the plasma peripheries. Departurs from LTE, i.e. the non-LTE (NLTE) state of the plasma, are represented by the deviation between the heavy-species (atoms, ions, molecultes) temperature T_h and the electron temperature T_e closer to the anode in Fig. 1 (*left*). The condition of quasi-neutrality, i.e. the balance between the numbers of positively and negatively charged particles, and the establishment of a composition following mass-action laws (e.g. Saha's equation) are also violated near the anode. Such deviations lead to drastic changes in the distribution of electron number density n_e and of electric potential ϕ.

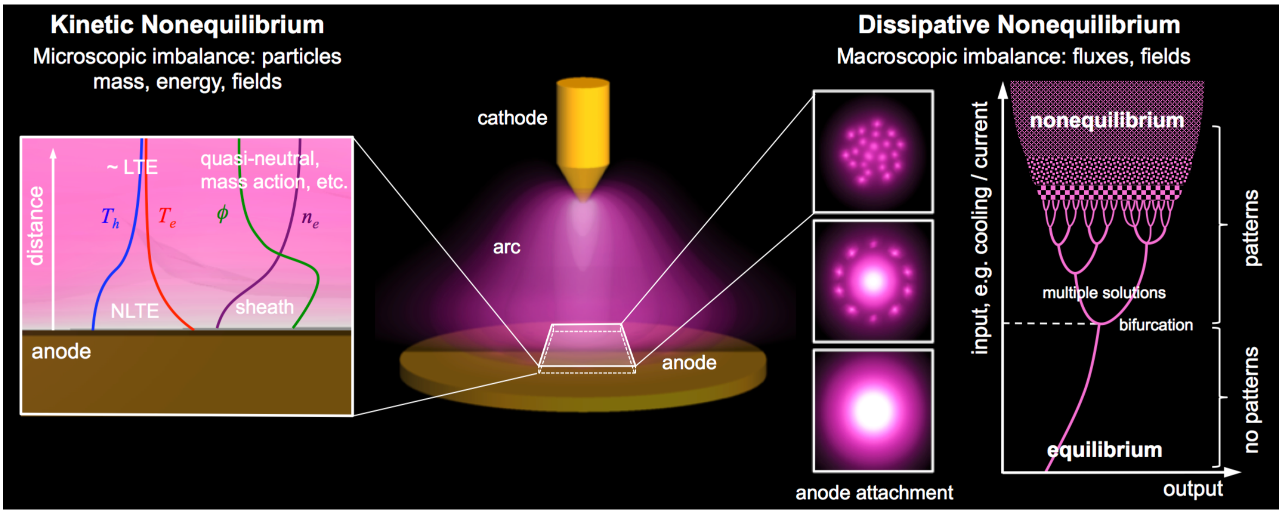

Fig. 1. Nonequilibrium in thermal plasmas: Kinetic nonequilibrium is the result of microscopic particle and field (particle kinetics) imbalances, which lead to deviations in distribution functions, mass action laws, quasi-neutrality, etc.. Kinetic nonequilibrium is exemplarily dominant in the region near the anode, manifested as the formation of the so-called *anode sheath*. Dissipative nonequilibrium is the result of imbalances among macroscopic fluxes and fields, which lead to the occurrence of multiple configurations (solutions) after surpasing a critical value of a controlling parameter (e.g. total current) at which a *bifurcation* occurs. These configurations achieve different degrees of energy dissipation, and are therefore known as *dissipative structures*. Dissipative nonequilibrium is exemplarily depicted by the formation of patterned property distributions along the anode surface for high degrees of anode cooling and low currents.

The description of kinetic nonequilibrium is directly related to the fidelity of the mathematical/physical model, and hence to the number of variables involved. For example, if LTE is assumed, only one variable, the equilibrium temperature T, can be used to describe the distribution of energy throughout the system; in contrast, under NLTE, two variables are needed, i.e. T_e and T_h, which characterize the energy carried by free electrons and heavy-species, respectively. Generally, the greater the degree of kinetic nonequilibrium captured by the model, the greater the model's fidelity, and the greater the umber of independent variables involved.

Representative types of kinetic nonequilibrium of relevance to thermal plasma flows are: (1) chemical nonequilibrium, of major relevance when complex molecular gas mixtures are used, in chemical synthesis processes, and when metal vapors are present; (2) thermodynamic nonequilibrium (i.e. NLTE), caused by the plasma - processing media interactions, such as a gas stream, electrodes, or feedstock material, as described above; (3) multi-phase interactions, as found near the plasma-electrode and solid material interfaces, as well as in novel processes such as liquid and vapor spraying; and (4) radiative transport, of major relevance during high-power operation, and particularly complex for molecular gas mixtures and high degree of nonequilibrium. The computational descriptions of these phenomena represent significant challenges in terms of model fidelity and are characteristic of multiphysics models. Strategies for multiphysics model involve different methods for model coupling, hybrid solvers, and multi-scale time stepping.

3. Dissipative nonequilibrium

Dissipative nonequilibrium is the result of imbalances among macroscopic fluxes, usually (but not necessarily) driven by the imposition of external constraints (e.g. mass flow, heat flux, applied voltage). Dissipative nonequilibrium is manifested as the establishment of competing configurations (i.e. multiple solutions) of the system, which appear when a critical value of a given controlling parameter P (e.g. total current, degee of anode cooling) is surpassed [3]. When the critical value of the control parameter P_{crit} is achieved, the system is prone to experience a *bifurcation* event in which two (or more) solutions co-exist. The system is in a dissipative equilibrium state for $P < P_{crit}$, and in a state of dissipative nonequilibrium for for $P > P_{crit}$. The degree of nonequilibrium becomes more pronounced the greater the value of P beyong P_{crit}. The multiple configurations of the system achieve different degrees of energy dissipation, and are therefore known as *dissipative structures* [3, 4]. The variety of solutions, and hence of dissipative structures, increases with the degree of nonequilibrium. For high dissipative nonequilibrium, the system is prone to a wide variety of configurations, and is generally considered as chaotic. For even higher dissipative nonequilibrium, the system achieves a turbulent state - the ultimate level of macroscopic dissipation.

A distinct example of dissipative nonequilibrium in a thermal plasma system is the self-organization of patterned property distributions along the anode surface – as depicted for the free-burning arc, a canonical arc discharge, in Fig. 1 (*right*). The distribution of T_h over the anode experiences the spontaneous (from a smooth axisymmetric distribution) formation of patterned configurations after a critical level of anode cooling [5]. The obtained patterns are also function of the total impossed current. For high current levels, the patterns have a planetary configuration, with a central spot and small spots along its periphery, that is also static. For higher degrees of cooling and lower current values, the distribution of T_h is composed of numerous spot patterns, which exhibit dynamic behavior. The computational results reported in [5] show qualitative agreement with experimental observations [6]. The progression of dissipative nonequilibrium states is not exclusive of the plasma – anode interaction; similar progressions can be expected in thermal plasma flows driven by strong interactions, as hinted by the computational results for the arc in cross inflow reported in [7] and those for the flow in a nontransferred arc plasma torch in [8].

Generally, the greater the degree of dissipative nonequilibrium to be captured by the model, the greater the numerical accuracy needed, and the greater the umber of discrete variables involved. Also generally, the greater the state of dissipative nonequilibrium of the system, the more multiscale the system is. Representative types of dissipative nonequilibrium phenomena of relevance to thermal plasma flows are: (1) flow stability, including fluid dynamic, thermal, and electromagnetic, often originated at the plasma periphery and leading to macroscopic flow reconfigurations; (2) pattern formation and self-organization, especially encountered in plasma-electrode interactions (as discussed above), which can cause enhanced erosion and process non-uniformities; (3) turbulence, the paramount example of multiscale phenomena and a major driver of gas entrainment and mixing; and (4) complex spatial and temporal configurations, as often found in modern plasma sources, such as multi-electrode or alternating power torches.

4. Conclusion

The unique characteristics of thermal plasma, amply exploited in applications, make them prone to exhibit nonequilibrium phenomena. Nonequilibrium in thermal plasmas, as in other multiphysics and multiscale systems, can be characterized as *kinetic* or *dissipative*. Kinetic nonequilibrium is a consequence of microscopic imbalances, as given by particle kinetics. Dissipative nonequilibrium results from macroscopic imbalances, often due to the imposition of external constraints onto the system. The computational description of kinetic nonequilibrium is directly related to the level of *fidelity* of the mathematical/physical model, whereas the description of dissipative nonequilibrium, to the level of *accuracy* of the numerical solution. The need for increased fidelity and accuracy can be potentially unbounded, whereas computational resources are necessarily bounded. This dichotomy implies that the modeler often faces the choice between performing "better" simulations (e.g. higher dimensionality, higher resolution) or using "better" models (e.g. greater span of phenomena accounted for, fewer modeling assumptions). Despite th great progress has been achieved in the computational modeling and simulation of thermal plasma flows, significanly more progress is needed in terms of both, fidelity and accuracy, to achieve models capable to concurrently describe kinetic and dissipative phenomena, especially within the context of industrial thermal plasma applications.

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