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# PRECURSOR ZONE OF AN ACOUSTIC SHOCK WAVE IN WEAKLY IONIZED GAS

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Several agents emerging from an acoustic shock wave modify its precursor zone with respect to free stream conditions. These are fluxes of charged particles accelerated or reflected from the potential barrier in double electric layer, radiation from relaxation zone, and ion-acoustic waves. Our study attempts to provide an insight into the processes initiated by these agents and to identify the character and the extent of the modification. We present a quantitative discussion of ion flux penetration in the free stream region, ionization-recombination process, and transfer of radiation from shock layer and relaxation zone to precursor zone. The condition in precursor region thus prepared becomes the initial condition for the processes in the relaxation region. Our model of recombination process is based on a non-steady state particle balance. Using this multiple-step approach of the combined action of several mechanisms, we will demonstrate how several observed effects of dispersion and propagation of shock waves in weakly ionized gas can be explained.

## Introduction

An accepted explanation of the anomalous dispersion and propagation of acoustic shock waves in a weakly ionized gas (WIG) has not yet been established despite a significant experimental and analytical effort to study possible physical mechanisms underlying the phenomenon. Debate continues about whether the kinetics of charged particles play an essential role, or whether the observed phenomena have instead a purely thermal origin.

Results from several recent experiments in shock-discharge tubes can be explained only by the existence of multiple electric layers across the shock structure [1]. Models that include even limited dynamics of charged particles indicate the existence of a very high-density space charge layer inside the shock structure and a strong "body force" induced by this space charge [2]. A model of a pulsed discharge crossing a normal shock indicates the possibility of strong back flow of gas particles into the free stream as a result of the interaction between the discharge and the shock [3].

Recent work suggests the existence of filamentary discharges perpendicular to the shock wave. Although there is not yet a direct experimental observation, the stability of these "unipolar" discharges has been confirmed [4]. The possibility that the charge separation region causes an unsteady region of transient, hot filaments suggests a way to transform electrostatic energy into heat, thereby coupling the charged and neutral particles. This opens a new possibility for explaining the anomalous effects.

We consider the following scenario as a plausible framework for our future analysis. A shock wave propagating through a weakly ionized gas induces a region of charge separation due to the difference in mobility between electrons and ions. This initial perturbation leads to the accumulation of high-density space charge at the shock front. This layer acts as a conductor and a breakdown can occur between the space charge layer and the free stream plasma. Breakdown waves propagate rapidly by a photo-ionization mechanism along many thin filaments, which are heated locally to temperatures comparable to the electron temperature. The shock wave propagates faster along these filaments and much slower along the colder freestream regions unaffected by the filaments. These filaments have transient, pulsed character because they initiate, expand, and fade away on a short time scale. However, while some filament decay, new ones form at different points of the front surface. The front end of each filament radiates and emits fluxes of particles into the free stream, preparing the precursor region for ionization by exciting the neutral particles, heating electrons, and generating local ion-acoustic waves. Shock dispersion occurs in this zone of filamentary discharges. Upon exiting the ionized gas region, the mechanisms of dispersion stop, and the shock wave resumes its original shape and continues to propagate with the normal velocity through the cold neutral gas.

Assuming this hypothetical qualitative phenomenology, we focus here to the shock precursor region, that is the medium, which has to sustain the chain of events described above. Our analysis shows that this region is substantially modified by the upcoming shock wave.

**Ionization Processes in Precursor Region**

In the non-equilibrium presentation of ionization process in the precursor region of a rare-gas discharge, the ionization rate for non-associative ionization contains three terms: direct ionization from the ground state

$$A + e = A^+ + 2e, \qquad (1)$$

ionization from an excited state

$$A^* + e = A^+ + 2e, \qquad (2)$$

and ionization in the collision of two excited states

$$A^* + A^* = A^+ + e. \qquad (3)$$

Direct ionization from ground state is important at very low pressure, usually below the range of interest for the present study. Ionization from an excited state is important process at higher pressure. There is a wide intermediary range of pressures where ionization by excited pair collision is important. This mechanism is responsible for the well-known Penning ionization in the case of gas mixtures or in the presence of impurities.

Other important ionization processes include ion conversion by termolecular associative ionization

$$A^+ + A + A = A_2^+ + A + e. \qquad (4)$$

Parallel to the energy-pooling process is the process of homonuclear associative ionization, also known as Hornbeck-Molnar process

$$A^* + A = A_2^+ + e. \qquad (5)$$

This process probably was undistinguished in the early experiments that still serve as the source of data. Hornbeck-Molnar process is possible only for excited atoms, which have excitation energy equal or larger than atom's ionization energy reduced by the dissociation energy of the molecular ion.

Rate coefficients for direct ionization from ground state (1) and for the ionization from the excited states (2) are available [5]. Their reliability depends on the reliability of cross section data available for the particular system. However, rate coefficients for the energy-pooling reactions are very poorly known. In the case of argon, which is one of the simplest target, all data cited in the literature can be traced to an estimate of a cross section for helium [6]. Based on that information and assumption that the rate coefficient should be proportional to thermal velocity, we estimated energy pooling rate coefficient, $k_p$ to be

$$k_p = 6.4\ 10^{-10}\ (T/300)^{1/2}\ cm^3sec^{-1}. \qquad (6)$$

Termolecular associative ionization is a relatively well known process [7]. It seems that simple power relationship $k_{AI} \sim T^{-n}$ is an accurate description in argon for the decrease of rate coefficient with heavy-particle temperature. As outlined in Ref. [7] simple estimate, based on the polarization interaction only, suggests n=0.75. However, derived empirical relation [8] based on several experiments performed in a wide temperature range, gives n=1.05. This result suggests that exchange interaction plays a certain role in this process. Extrapolating these data to the gas temperature range which is characteristic of the precursor region, indicates factor of two discrepancy between the simple estimate [7] and empirical data [8-10], which is presented in Fig. 1. Therefore we adopted the following relation

$$k_{AI} = 2.4\ 10^{-31}\ (T/300)^{-1.05}\ cm^6sec^{-1}. \qquad (7)$$

In Hornbeck-Molnar process, providing that the excited atoms are in thermal equilibrium with free electrons, the rate coefficient should be dependent on both, electron and ion temperatures [11]. Extensive review of this process was given Ref. [12]. The result of this analysis is justified by the experimental result obtained by Johnson and Gerardo [13] for the typical conditions of a pulsed discharge induced by an electron beam, which is $k_{HMI} = 3.5\ 10^{-10}\ cm^3sec^{-1}$. Since the threshold for this process in argon is about 14.6 eV, which is about 0.5 eV deeper than in the case of xenon, the expression for rate coefficient in argon should be

$$K_{HMI} = 5.9\ 10^{-10}\ T^{1/2}\ \exp(-2.1/kT_e) \quad (8)$$

in the units of $cm^3sec^{-1}$. A relatively simple theory developed by Janev and Mihajlov [14, 15] was used in alkali plasmas to estimate rate constants for Hornbeck-Molnar process.

Relative contributions of the reactions described by Eqs. (1-5) to the ionization process depend on the plasma parameters, gas composition and flow characteristics. Therefore, there is a variety of possible ionization paths. For instance, influence of molecular ions in rare gases is negligible at pressures below 1 Torr, but dominant at pressures above 10 Torr and at low degree of ionization. It is negligible again at high electron density and high pressures. Strong dependence of termolecular associative ionization on gas temperature creates substantial effect in cold discharges, and moderate transient effects in strong ionizing waves [16]. Hornbeck-Molnar process is effective at both, high electron and high gas temperatures.

In typical conditions for shock tube WIG experiments, the dominant molecular ions are $Ar_2^+$ in argon, $N_4^+$ in nitrogen, or $NO^+H_2O$ in air with traces of water. These ions are heavier than those that normally occur in thermal plasmas. This makes space charge effects even more pronounced and the hypothesis of a narrow, highly conductive positively charged layer around the original shock front seems quite plausible. Further, the dominance of these heavy molecular ions means that the degree of ionization is effectively increased. Finally, their relatively long lifetimes, decay paths into metastable states combined with the mutually ionizing collisions of atomic metastable states are probably responsible for long-lasting effects in the afterglow [17]. On the other hand, molecular ions are not dominant in pulsed discharges [18] due to the presence of non-thermalized electrons, and rapid recombination effects.

In order to treat quantitatively the ionization processes involving excited states a separate model was introduced [19]. Transient population of a number of aggregated representative excited states was obtained by solving simultaneously a system of equations for excited state populations. The system describes temporary balance of excited states by combining gains by excitation, de-excitation from upper levels, and recombination with the losses by ionization or de-excitation to lower levels. Collisional-radiative recombination rate coefficient for non-associative ionization processes is taken from Ref. [20].

Solving a system of non-steady state equations using the available rate coefficients derived distribution of atomic, molecular, and ionic species. The system was converted into position-dependent by transformation in the coordinate system traveling with the shock wave. The results were "frozen" population distributions across the precursor region. Practically, the excited states were grouped into a representative set of states thus reducing the number of equations involved.

**Energy Fluxes**

The electric field immediately after the shock front may increase to the levels of kV/cm (illustrated in Fig. 2) and to drop to about 10 V/cm in most of the relaxation zone. There is a field reversal at the peak of the electron cascade front. The remainder of equilibrium zone has a low negative electric field of the order of 1 V/cm mostly due to the cooling effects. This field is superimposed to the electric field of discharge's positive column. Hence, there are the observable difference in temporal behavior of voltage and current when shock wave propagates from cathode to anode and opposite [1]. The induction of sizable electric field across the precursor region is reflected in substantial counter-flows of charged particles. These charged particle fluxes usually do not carry enough energy to modify substantially precursor region. The levels of reduced electric field, E/N are typically less than 30 Td (see Fig. 2). Therefore, while the ion counter-flow is not expected to have any influence, either on the shock structure or on the build-up of excited-state population in the precursor region, it may serve as the simmer current for breakdown of filamentary discharges.

Conditions for ion acoustic instabilities are fulfilled in plasma parameter range characterizing the precursor region. It is known that in a shock tube experiments electron drift velocity is higher than ion acoustic wave velocity, but lower than electron thermal velocity. Thus, the ion acoustic instabilities should be generated around the electric double layers. It is the extent of ion acoustic wave propagation, as well as the effect of its damping in the precursor region through local changes of

electron heat conductivity that deserves more attention. The analysis of ion-acoustic waves in the precursor region is further complicated by the existence of multiple ion species, hence of the multiple ion propagation modes. Problems of propagation of ion acoustic waves through precursor region should not be *a priori* dismissed as irrelevant. It deserves more attention.

The last remaining energy flux, absorption and diffusion of resonant radiation can penetrate through the translational shock into precursor region. Resonant radiation from denser shock layer propagates to more dilute precursor region. According to the Biberman-Holstein theory [21, 22] radiation trapping occurs with partial frequency redistribution. The situation is similar to radiation transfer in the boundary layers of a glow discharge, where the number of photo-excited atoms and molecules may be given by

$$\frac{dN_{exc}}{dt} = -A(N)N_{exc}, \qquad (9)$$

where A(N) is the density-dependent trapped radiation decay rate. Transforming the relation into the one-dimensional frozen shock presentation, we obtain

$$\frac{dN_{exc}}{dx} = -A(N(x))N_{exc}. \qquad (10)$$

Using a simple analytical formula [23] for radiation trapping with partial frequency distribution one can estimate the effect of absorption of resonant radiation on the population of first resonant level in the precursor region. The effect is illustrated in Fig. 3. Results of conventional models based on the thermal equilibrium (TE) with ground state or TE with free electrons can not reproduce the excited-state population profile. Behind the translational shock and in the precursor region this profile was reproduced both, qualitatively and quantitatively with the present model, which incorporated the absorption of resonant radiation.

## Conclusion

In this paper we outlined the issues that has to be addressed in order to properly characterize the precursor region in WIG flow models. Although there are still many "blank spots" in our knowledge of appropriate parameters, the region can be quantitatively characterized and more accurate initial conditions for the models can be obtained. Composition of ions and excited states may vary substantially in the range of parameters found in shock-WIG problems.

We underlined the fact that heavy ions may be abundant or dominant in the precursor region of the shock wave. In addition to complex ion and excited state composition, energy fluxes from the shock may modify substantially the precursor region. Radiative transfer with partial frequency distribution, nonlinear damping and interaction of multiple-mode acoustic waves, and charged particle fluxes are effective in this region.

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