Negative Mobilities of Electrons in Radio Frequency Fields

Saša Dujko, Zoran M. Raspopović, Zoran Lj. Petrović, and Toshiaki Makabe

***Abstract*—In this paper, we perform calculations of mobilities (represented by drift velocities) of electrons in mixtures of F_2 and Ar in radio frequency (RF) fields. The conditions were chosen which correspond to those that led to observation of negative absolute mobility in dc fields in decaying plasmas. In RF fields, a similar effect is observed as the mobility corresponding to the flux drift velocity is negative. At the same time, it was found that the mobility corresponding to the bulk drift velocity is mainly positive in respect to the expected value, but that it has a large phase delay. The effect is caused by nonconservative nature of the attachment and in the absence of nonconservative collisions both mobilities are positive and in phase with the field.**

***Index Terms*—Drift velocity, electron transport, nonequilibrium, radio frequency (RF) plasmas.**

I. Introduction

In RECENT years, attention has been paid both to theoretical studies of negative electron mobility in low-temperature plasma and to associated kinetic phenomena. The negative mobility (conductivity) may be first associated with the negative differential conductivity (NDC) that was explained on the basis of kinetics of energy and momentum relaxation [1]. A possible connection comes from the fact that both effects appear to be favored by the increasing momentum transfer cross section and also by the fact that in the low-field limit, the mobility is positive and, therefore, some form of NDC is required to give rise to a possibility of a negative mobility.

The most important features of NDC in radio frequency (RF) fields using Monte Carlo (MC) simulations were discussed in [2]. At the same time, it was shown by using momentum transfer theory (MTT) [3] that nonconservative collisions may induce NDC by two different mechanisms, the first through the effect on the electron energy distribution function (EEDF) and the second through the effect on the bulk drift velocity.

Another even more closely related physical situation is the transient negative conductivity (TNC). TNC was observed experimentally in time-resolved conductivity measurements in relaxing Xe plasma, ionized by a hard X-ray pulse [4] or by laser radiation [5], and explained theoretically [6]. This nonequilibrium kinetic phenomenon is a result of a strong dependence of electron collision frequency on electron velocity and it is characteristic of electrons with an initial mean energy in the region of rapidly rising cross section such as the region above the Ramsauer–Townsend minimum.

The possibility of negative conductivity under steady-state conditions in beam sustained plasma in gas mixtures with a small amount of electronegative gas is another similar physical situation. Several gas mixtures have been studied: Ar:CCl_4, Ar:F_2 [8], and Xe:F_2. Unfortunaly, there are no experiments for these mixtures under steady-state conditions. Another physical situation giving a possibility of negative conductivity is in Ar:Li and Ar:Li:N_2 photoplasma and it has been theoretically studied in [9].

Finally, a possibility of negative conductivity has been discussed for Ar:NF_3 plasma under steady-state Townsend conditions [10] and in Ar:NF_3 [11] and Ar:F_2 [11]–[13] gas mixtures under afterglow conditions, where plasma would decay in a very weak electric field. In the case of decaying plasma under afterglow conditions, it was predicted by using two different methods, MC simulations and the solution of the Boltzmann equation, that a stationary or quasi-stationary negative mobility of electrons may be achieved [13]. The formal explanation of this kinetic phenomenon is based on the fact that attachment removes thermalized electrons from the plasma, thus heating the electron swarm or burning a hole in the low energy part of EEDF. It is quite obvious that the key mechanism of this effect is the nonconservative nature of the electron attachment. In addition, attachment at low energies provides a stationary character of the effect otherwise it would be transient. As a result of strong attachment that gives rise to the negative electron mobility, which at first appears to be in contradiction with the basic laws of thermodynamics, the whole system, i.e., plasma, decays rapidly, even though the transport coefficients have reached stationary values.

The primary goal of this work is to study the possibility of development of the negative mobility (NM) by using RF fields. The basic idea is to test how this effect would develop in RF fields as one may be able to provide circumstances where one would have a certain part of the period when ionization may be sufficient to compensate for the decay of plasma and, thus, to achieve a practical implementation. Another option would be to use the spatially distinct regions of higher current RF plasma, the edge of the sheaths for ionization and then to achieve negative conductivity in the bulk where field is relatively low. In addition, a number of interesting kinetic phenomena that have been observed for RF plasmas gave an idea that in studying this example one may provide a new insight into either electron transport in RF fields or into the negative mobility itself.

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MC simulation procedure that has been developed for time varying fields and tested on a wide range of examples [14] was employed in this paper. We have selected the mixture of Ar-F_2 under conditions, which led to negative electron mobility in the afterglow—dc field conditions. On the other hand, this mixture corresponds to the gas composition generally used in plasma processing since most of the active gases for dry etching are electronegative.

II. MC Procedure

The electron swarm is assumed to develop in an infinite gas under uniform fields. The boundary effects are neglected as well. As these calculations are made for swarm conditions, it is assumed that the electron density is sufficiently small so Coulomb interactions between the electrons as well as shielding of the field are negligible. In addition, the gas is not perturbed from thermal equilibrium by the presence of the electron swarm.

We have followed a large number of electrons, typically 10^4–10^6 over small time steps to reduce the statistical uncertainty of MC simulation since the negative mobility was shown to be relatively small in case of dc fields. Our MC code is based on time-integration technique where small time steps are made to move the electrons and update the value of the field. The duration of the steps is determined by the minimum of the three relevant time constants (mean collision time, period of the field and cyclotron period for $\boldsymbol{E} \times \boldsymbol{B}$ fields if magnetic fields are present) divided by a large number (20–100). Period of the electric field is always divided by at least 100, and these moments are used to sample the transport properties.

When attachment is a significant process, such as in the case studied here, or when ionization is present, the numbers of electrons were scaled up or scaled down, respectively, during the simulation. This is equivalent to having a constant collision-frequency ionization or attachment and in a separate simulation this procedure was shown to give correct results [13]. In the present gas mixture, a very large attachment cross section occurring at low energies leads to a very nonconservative nature of electron transport. In order to realize correct representation of nonconservative electron transport, we have used the formulae to sample transport coefficients from [15] and [16]. Thus, we have calculated two types of transport coefficients, the ***bulk*** and the ***flux***. The *bulk* drift velocity is defined as

$$\vec{v}_b = \frac{d}{dt}\sum_{i=1}^{n}\vec{r}_i \tag{1}$$

and the *flux* drift velocity as

$$\vec{v}_f = \sum_{i=1}^{n}\vec{v}_i \tag{2}$$

where n is the number of electrons and r_i and v_i are the position and velocity of the ith electron.

It follows from (1) that bulk drift velocity is displacement of the mean position of the electron swarm and it characterizes the motion of the total ensemble of electrons. The *flux* drift velocity (2.2) is the mean velocity of electrons. In the case of uniform plasma, the *flux* drift velocity corresponds to the drift velocity obtained from Boltzmann equation analysis. In general, there are no experiments for measuring *flux* transport coefficients, but in most aspects of plasma modeling, these transport properties are required. Therefore, MC simulations have a great advantage related to swarm experiments as one would be able to make the distinction and point out how different those two are. In plasma modeling, it is usually assumed that experimental data may be employed but any example giving a significant discrepancy between *flux* and *bulk* properties may be a warning for plasma modelers. *Bulk* and *flux* drift velocities are the same under conservative conditions.

III. Results and Discussion

A. Description of Gas Mixture and Conditions of Simulation

Physical object of MC simulation is electron swarm under influence of electric and magnetic field. We have performed simulations for various conditions but the following will be labeled as "standard conditions."

- Gas mixture: (0.5%) F_2/Ar.
- Gas temperature: 0 K.
- Gas number density: $\mathrm{N} = 0.24 \cdot 10^{26}\ \mathrm{m}^{-3}$.
- Electric field time dependence: $\mathrm{E(t)/N} = 0.141 \cdot \cos(2\pi \mathrm{ft})$ Td.
- Frequency of applied electric field: $\mathrm{f} = 200$ MHz.
- Initial electron energy distribution is Maxwellian with the mean energy of 1 eV.

A set of cross sections for electron scattering on Ar and F_2 used in our MC simulations is the same as given in [13].

B. Relaxation of Mean Energy and Transport Coefficients

Relaxation of the RF electron swarm is the starting point of our study. Relaxation itself is an example of transitive regime into final, quasi-stationary regime, that is inherently nonequilibrium. Relaxation of transport coefficients in RF fields is complex and sometimes difficult to predict on the basis of dc theory [17]. In addition, the actual plasma models such as the relaxation continuum theory (RCT) may require knowledge of the realistic relaxation times in order to describe the temporal (and spatial) development of electron transport coefficients in RF plasma models [18].

In RF fields, relaxation of transport coefficients will depend on whether those are associated with momentum or energy balance. The difference in achieving either of the two balances at different conditions (frequency, pressure, and field) will lead to a number of kinetic phenomena such as anomalous diffusion [19]–[21], RF negative differential conductivity [2], [22], [23] (for reviews, see [24] and [25]). While it is of interest to be aware of the temporal relaxation in order to understand kinetic phenomena in swarm transport, it is even of greater importance to understand it in order to be able to interpret even more complex phenomena in gas discharges [26]–[28].

In Fig. 1, we show the temporal relaxation of the mean energy of electrons for pure argon and for various gas mixtures of Ar–F_2, otherwise under standard conditions. As can be seen, relaxation time for pure argon is the largest while increasing abundance of F_2 in gas mixture leads to decreasing of relaxation

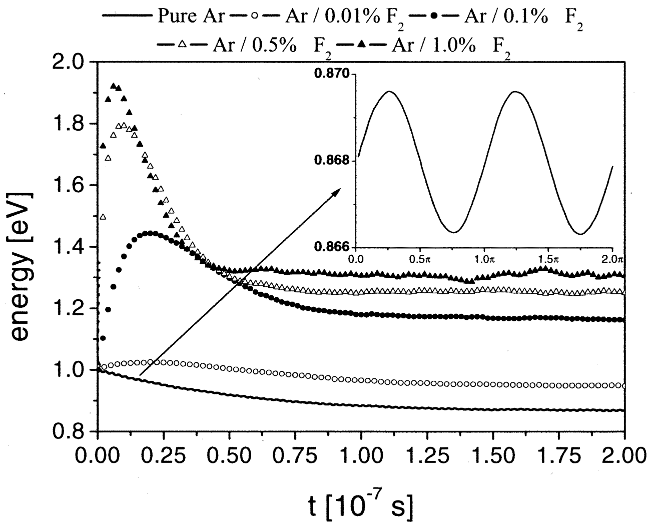

Fig. 1. Temporal relaxation of the mean energy of electrons for different gas mixtures under standard conditions.

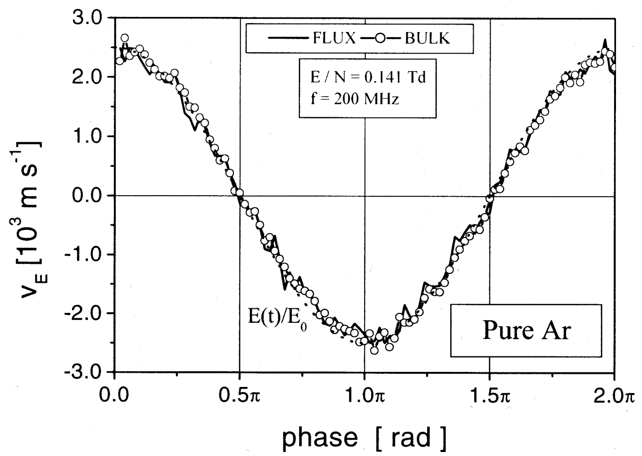

Fig. 2. Time dependence of drift velocities in pure argon under standard conditions. Subscript "E" denotes direction of the field.

time as can be expected for a mixture of atomic and molecular gas. However, the increasing F_2 abundance leads to increasing of the mean energy which is in contradiction with the previous conclusion and is, thus, a clear sign of a pronounced attachment heating. As one can see from Fig. 1, the modulation of the mean energy due to variation of the field is small, almost impossible to observe, which shows that the influence of inelastic collisions under present conditions is relatively small. Similar relaxation behavior is observed for all transport coefficients except that they are modulated according to the field direction.

C. Quasi-stationary Drift Velocities

Once relaxation has been completed, the time dependence of transport coefficients reveals the relaxation due to the variation of the electric field but not due to initial conditions. Our main interest is in drift velocities and in Fig. 2, we show the *flux* and *bulk* drift velocities obtained in pure argon over one period of the electric field. As we mentioned previously, the lack of non-conservative collisions at such low E/N leads to situation where

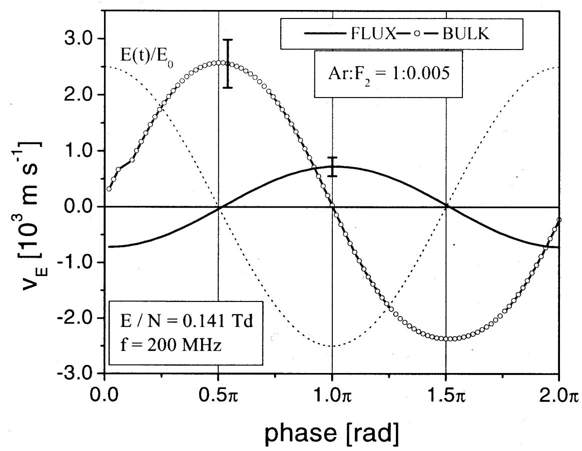

Fig. 3. Time dependence of drift velocities in Ar–F_2 gas mixture under standard conditions. Dotted line indicates, in this and all subsequent figures, the expected direction of drift velocity that follows the electric field and corresponds to a positive mobility. The data presented here have been smoothed for clarity and the maximum scatter is shown by error bars.

the *flux* and *bulk* drift velocity are identical and the results of the MCS are in excellent agreement, thus showing internal consistency of calculation. One should note that we denote the axis along the direction of electric field by using subscript "E." In addition, one should bear in mind that both drift velocities are presented as positive if their mobility is positive. Also, the phase differences of drift velocities and the field, which has a cosine time dependence, are negligible.

When we introduce the fluorine into the gas mixture, a completely different situation occurs, as shown in Fig. 3. The *flux* drift velocity is exactly in the opposite phase to the electric field with a very small or negligible phase shift while the *bulk* drift velocity has a large phase shift. Therefore, the mobility corresponding to the *flux* drift velocity is negative while the one related to the *bulk* drift velocity is mainly positive.

In an attempt to explain the observed situation, we should remind ourselves of the physical explanation of the negative mobility and of the difference between the *bulk* and *flux* drift velocities under the circumstances leading to negative mobility. Negative absolute mobility is result of the fact that electrons that are being thermalized in the region of rapidly rising momentum transfer cross section have a greater chance of collision if they move in the direction opposite to the field and, thus, gain energy as compared to the electrons that move in the direction of the field which lose their energy. The latter group becomes dominant due to increased scattering of the former group and it gives rise to the negative mobility which would under normal circumstance be only transient [13]. However, if there is a low-energy attachment, the second group of electrons which would eventually stop and be accelerated by the field, disappears and the effect becomes stationary, albeit with a loss of electrons due to attachment. The same explanation appears to be valid in the case of RF fields for the flux drift velocity. One does not expect a significantly increased phase shift between *bulk* and *flux* drift velocities on the basis of standard relaxation. Yet, the large phaseshift that is observed corresponds well to the requirement that the conditions for attachment cease at one end of the swarm and have to be created at the other as the field changes direction.

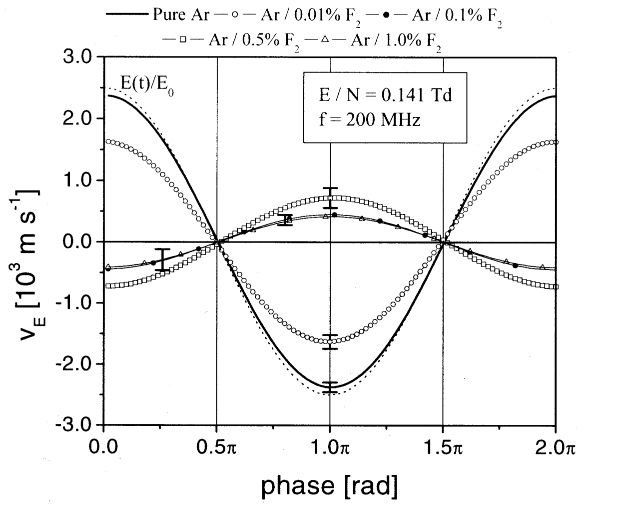

Fig. 4. Time dependence of the *flux* drift velocities for different gas mixtures under standard conditions. Dotted line shows the expected dependence based on positive mobility (the time dependence of the field), the error bars indicate the maximum scatter and the data for 0.1% and 1% of F_2 overlap very much.

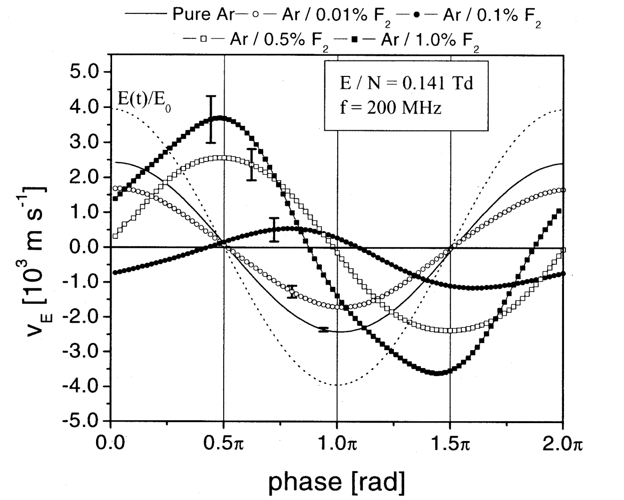

Fig. 5. Time dependence of the *bulk* drift velocities for different gas mixture under standard conditions. Dotted line shows the expected dependence based on positive mobility (the time dependence of the field), the error bars indicate the maximum scatter.

On the other hand, it was found that in dc fields the mobility related to *bulk* velocity is positive even when the mobility due to the *flux* velocity is negative [29]. This result was supported by thermodynamic argument on the basis of calculation of the total entropy where it was found that the second law of thermodynamic is not violated in case of NM if the entropy production due to attachment is included and the terms in entropy balance clearly reflect terms in calculation of the *bulk* drift velocity which, as a result, is always positive. The behavior of the *bulk* drift velocity was explained by proposing that, while majority of electrons are moving in the opposite direction to that which is normally expected, a wave of attachment is eating away those electrons [13], [29]. As the electrons are spatially separated, the low-energy electrons disappear due to attachment and, therefore, the center of mass is moving in the direction that is expected, that is, opposite to the motion of individual electrons. The same is true in RF fields and the phase shift of *bulk* drift velocity which is introduced actually consists of the time required to establish spatial separation and a new front of attachment. This picture is supported directly by the simulations that were performed in dc fields. On the other hand, the relaxation of the direction of motion is much faster and *flux* drift velocity changes sign very rapidly.

In Figs. 4 and 5, we show results for *flux* and *bulk* drift velocities, respectively, obtained for different gas mixtures under standard conditions. The results show that increasing the abundance of F_2 in gas mixture has a significant influence on the electron transport. Even at the abundance as low as 0.01% the *flux* drift velocity is significantly reduced while the phase difference in the *bulk* drift velocity appears. When the abundance of F_2 is increased further mobility related to the *flux* drift velocity becomes negative while the one associated with the *bulk* drift velocity changes its phase even further. The effects appear to saturate between 0.5% and 1% and then to decrease with further increase of the percentage of F_2.

Other transport and rate coefficient have normal behavior with the increase in the percentage of molecular (attaching) gas. For example, in Fig. 6, we show the time dependence of the attachment rate coefficient obtained for different gas mixtures under standard conditions. As expected, increasing the abundance of F_2 leads to larger attachment rates. One should also note that attachment rate coefficient shows very small modulations.

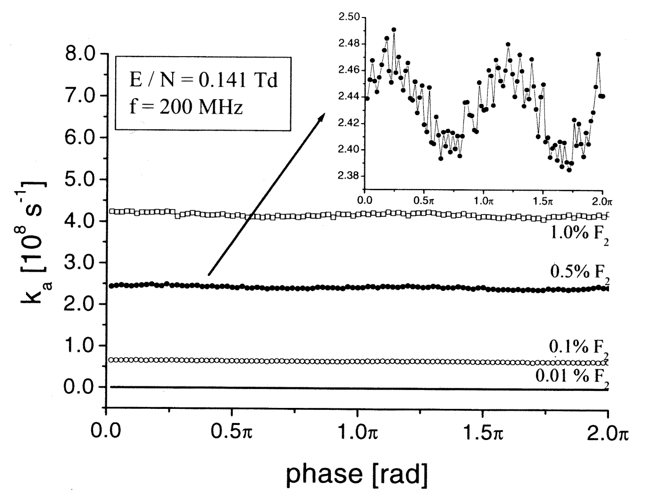

Fig. 6. Time dependence of the attachment rate coefficient for different gas mixtures under standard conditions.

D. Attachment Heating of RF Swarm

As we have already mentioned (see Fig. 1), an increasing abundance of F_2 leads to a higher mean energy of electrons. In order to understand the role of the attachment process, in our MC simulation we have changed the property of the attachment process into an energy loss-inelastic process (with the energy loss significantly smaller than the mean electron energy) with conserved number of electrons. In Fig. 7, we show the drift velocities for such conservative gas mixture. With the change in the nature of the process the drift velocities immediately turn to the expected behavior, with both positive mobilities and a very

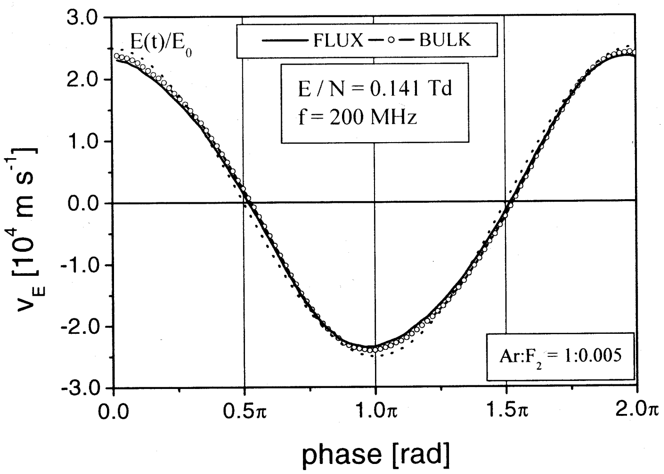

Fig. 7. Time dependence of the flux and bulk drift velocity under standard conditions with attachment treated as a conservative inelastic process.

small phase shift. This is direct proof that the nonconservative nature of the attachment is the basis of the negative mobility. The effect of attachment heating, as shown in Fig. 1, also disappears and the mean energy drops to around 0.2 eV with a slightly larger modulation due to artificial inelastic losses.

IV. Conclusion

In this paper, we discuss how negative absolute mobility as observed in dc afterglow plasma [13] is also observed in RF swarms. In addition, we show that the distinction between *bulk* and *flux* drift velocities is not just in the opposite directions but also in a large phase delay introduced for the *bulk* drift velocity. The flux velocity is required in most situations when plasma is modeled and one should bear in mind that a possibility that it may become slightly negative rather than positive will affect the calculated power balance. In addition, the cases where NM is observed are very similar to standard mixtures used for plasma etching and many other applications.

The key issue in discussion of the negative absolute mobility is whether it may be realized in practice and even yield some practical applications. The thermodynamic study of Robson *et al.* [29] actually points out that the total entropy of the system increases as evidenced by the positive bulk drift velocity. One should bear in mind that it is only the *bulk* drift velocities that are measured in standard swarm experiments. However, if some property depending on the *flux* drift velocity only can be identified then one may be able to point out a way to either observe the negative mobility or even to use it.

Another issue is whether the system would be useful at all. In the dc case studied so far in the literature, the price of negative mobility is pied by a rapid decay of plasma. In this paper, we propose a system, which may provide a possibility to actually achieve and apply NM. It is possible that during some parts of the period the field would be high enough to provide ionization and, thus, compensate for the losses of electrons in the other parts of the period. One option to achieve this would be to add the third component into gas mixture with a very low threshold for ionization (such as cesium vapor).

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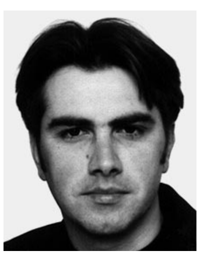

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