Negative differential conductivity of electrons in pure rare gases

N. L. Aleksandrov,[1] N. A. Dyatko,[2] I. V. Kochetov,[2] A. P. Napartovich,[2] and D. Lo[3]
[1]Department of Physical Mechanics, Moscow Institute of Physics and Technology, 141700 Dolgoprudny, Russia
[2]TRINITI, 142092 Troitsk, Moscow Region, Russia
[3]Physics Department, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong
(Received 2 May 1995)

A mechanism for negative differential conductivity (NDC) in pure rare gases is presented. The drift velocity of electrons in these gases has been calculated using the two-term expansion of the anisotropic electron energy distribution function in Legendre polynomials. In ionized rare gases (viz. Ar, Kr, and Xe), Coulombic collisions cause electron drift velocity to become a decreasing function of the electric field for specific energy levels and over a certain range of electric field strength. A detailed investigation of the effect of Coulomb interactions and electron collisions with excited atoms on NDC has been performed.

PACS number(s): 52.20.−j, 52.25.Dg, 52.25.Fi

I. INTRODUCTION

Negative differential conductivity (NDC) defined as the decrease in the drift velocity of electrons (or holes) with increasing electric field has been observed in weakly ionized gases [1,2] and in semiconductors [3]. This phenomenon induces spontaneous oscillations in externally sustained direct-current (dc) gas discharges [4–6], and is important for gas lasers [7,8] and devices detecting nuclear radiation [9–11]. Drift-tube measurements of the electron drift velocity in gaseous mixtures with NDC are used to normalize (scale) electron-molecule collision cross sections [12–14].

The occurrence of NDC has been explained in terms of the special features of the elastic and inelastic collision cross sections of the electron [15,16]. NDC can occur when the combination of the electron elastic and inelastic cross sections is such that an increase in the reduced electric field E/N (E is the electric field strength and N is the gas number density) leads to an abnormally large increase in the elastic collision frequency. The resulting enhanced randomization of the directions of the velocity vectors can decrease the drift velocity even though the mean electron energy increases. The combination of the rapidly increasing elastic cross section for Ar, Kr, Xe above the Ramsauer-Townsend minimum and the vibrational excitation cross section for some molecules is known to produce the NDC effect when the specific mole fraction of the molecule in the gas mixture (rare-gas atom and molecule) is small. Recently it has been shown that NDC may be observed in mixtures with helium (He) in place of molecular gas (i.e., rare-gas mixtures) [17,18]. In this case, the greater energy exchange in collision between electrons and He because the smaller mass of He plays an analogous role to weak inelastic collisions in molecule–rare-gas mixtures.

We present in this work a new mechanism for the occurrence of NDC, which may be observed in pure rare gases. This paper reports that results of calculations of the drift velocitv of electrons in Ar, Kr, and Xe predict that NDC can occur in these gases at certain values of degree of ionization when Coulomb collisions become important. At such values of degree of ionization, there exists in the plasma singly ionized atomic and molecular ions, the total number density of which is equal to the electron number density. We study the effect of the e-e (electron-electron) interaction and the collisions between electrons and excited atoms on NDC.

II. MODEL CALCULATIONS

We would like to point out that the rare-gas plasmas under consideration here are weakly ionized, meaning that the degree of ionization $\alpha = n/N$ (where n is the electron number density) is in the range from 10^{-9} to 10^{-4}. The physics in weakly ionized plasma differs greatly from that of plasmas that are fully ionized ($\alpha = 1$). The fundamental processes adopted in our model calculations are typical of those used in modeling weakly ionized plasmas.

Solving the Boltzmann equation with the two-term expansion of the electron distribution function in Legendre polynomials, we calculate the electron drift velocity, which can be written as [19]

$$w = -\frac{eE}{3N}\left[\frac{2}{m}\right]^{1/2}\int_0^\infty \frac{\varepsilon}{\sigma_m}\frac{\partial f_0}{\partial \varepsilon}\partial\varepsilon \ , \qquad (1)$$

where e and m are the electron charge and mass, respectively, σ_m is the electron-atom collision cross section for the momentum transfer, $f_0(\varepsilon)$ is the isotropic component of the electron energy distribution function (EEDF). The processes considered here are the e-e, e-i (electron-ion) elastic collisions, the elastic electron-atom scattering, the direct and stepwise excitation and ionization of atoms by electron impact. Any inelastic e-i collision may be neglected because of the low degree of ionization considered here.

The role of e-e collisions in the formation of the anisotropic component of the electron energy distribution function is implicit (see, for instance, [20]), because these collisions conserve the electron momentum. The energy exchange in *e*-*e* collisions is very effective, and its rate may be characterized by the effective cross section of the order of 10^{-12} cm^2 at energy equal to 1 eV. The *e*-*e* collisions exert an influence on the shape of the EEDF. The contribution from *e*-*e* collisions become comparable to electron-atom collisions when the degree of ionization $\alpha = n/N$ (where n is the electron number density) is about 10^{-9}.

The momentum transfer cross sections for Ar, Kr, Xe are taken from [21]. At high electron number densities, efficient excitation of atoms by electron impact can increase strongly the fraction of excited atoms. Therefore, we have also carried out a calculation taking into account collisions between electrons and atoms in the first metastable state including excitation and quenching of the excited atoms. The cross sections of interactions of electrons with excited Ar and Kr are taken from [22], and the data in [23] are used for Xe.

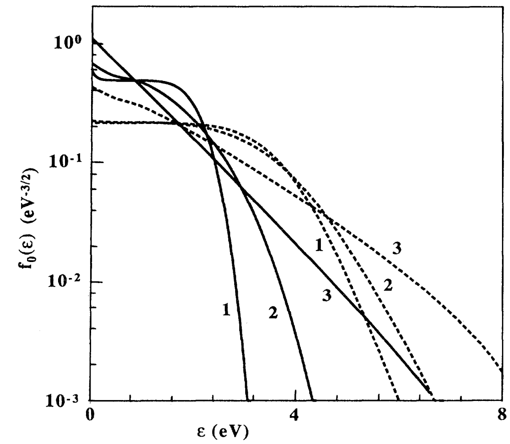

FIG. 2. Electron energy distribution functions in Xe at the following values of E/N in V cm^2: 4×10^{-18} (full curves); 2×10^{-17} (broken curves). (1) $\alpha=0$; (2) 10^{-7}; (3) 10^{-5}.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the calculated w in pure Xe versus E/N for several values of α and fractions of excited atoms $\delta = N^*/N$ (the excitation degree, and N^* is the number density of the atom in the first metastable state). The processes considered are collisions of electrons with electrons, ions, and ground-state and excited atoms. Collisions with an Xe atom in the first excited state may result in the excitation to the next excited state with a loss of electron energy of 1.16 eV, or the ionization or deexcitation to the ground state. NDC is clearly seen to occur at not too high and not too low degree of ionization. The results for pure Xe at $\delta=0$ are surprising since it was previously assumed that NDC occurs only for systems for which there exists some inelastic collision processes [2,15,16] or elastic collisions with light atoms (He) [17,18].

In order to explain the origin of the NDC effect in a pure rare gas with Coulomb collision, we show in Fig. 2 EEDF's in pure Xe for different δ and E/N for $\delta=0$. At low δ, collisional relaxation of electrons with energy corresponding to the Ramsuaer-Townsend minimum in the momentum transfer cross section is very low. Therefore, at not too high energies, the EEDF is almost constant. Electron-electron collisions are most effective at low electron energies because of the ε^{-2} dependence of the Coulomb collision cross section on energy. Hence *e*-*e* collisions drive the EEDF towards a Maxwellian at first in the low-energy region. Then this region where the EEDF is close to the Maxwellian grows with δ. This change of shape of EEDF is accompanied by diminishing mean energy (see Fig. 3) and the growth of w may be explained in two ways: as a result of an increase of the integrand $(-\partial f_0/\partial\varepsilon)$ in (1) or a decrease of the momentum transfer cross section at the mean energy (see Fig. 4). The NDC effect will take place if the increment in w is greater at the lower electric field. Numerical calculations

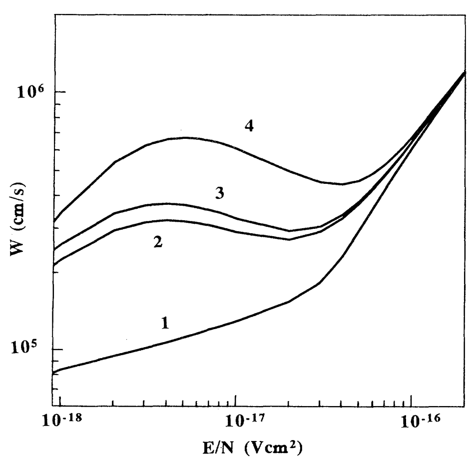

FIG. 1. Electron drift velocities in Xe vs E/N. (1) $\alpha=\delta=0$; (2) $\alpha=10^{-6}$ and $\delta=0$; (3) $\alpha=\delta=10^{-6}$; (4) $\alpha=10^{-6}$ and $\delta=10^{-5}$.

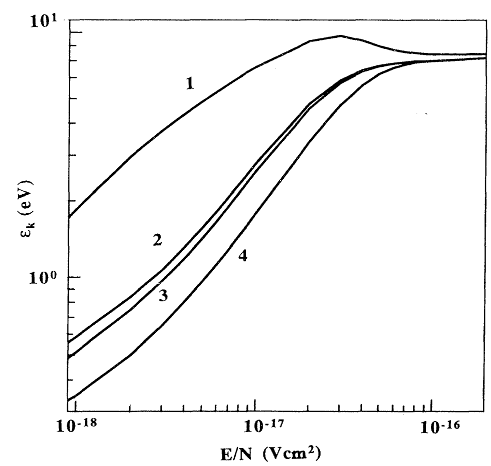

FIG. 3. Characteristic energy ε_k vs E/N for conditions of Fig. 1. $\varepsilon_k = D/\mu$, D is the electron transverse diffusion coefficient, μ is the electron mobility.

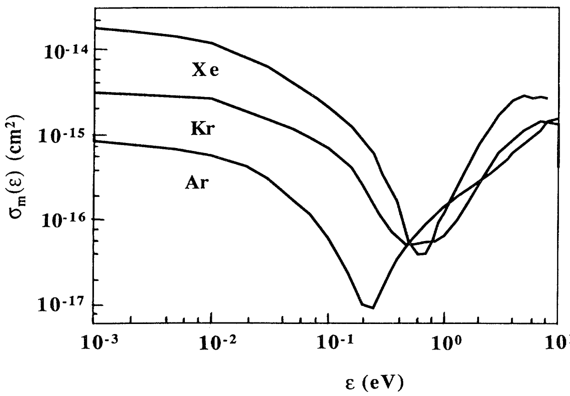

FIG. 4. Momentum transfer cross sections vs electron energy for Ar, Kr, Xe.

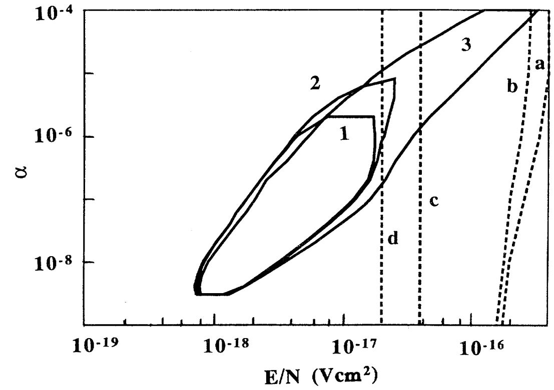

FIG. 5. $\alpha - E/N$ diagram for Xe. Full lines: bound region where the NDC exists. (1) $\delta = 0$; (2) $\delta = \alpha$; (3) $\delta = 10\alpha$. Broken lines correspond to ionization-recombination balance. (a) $\delta = 0$; (b) $\delta = 4\alpha$; (c) $\delta = 10\alpha$; (d) $\delta = 100\alpha$.

(Fig. 1) demonstrate that this is the case. For a high degree of ionization, $\alpha = 10^{-5}$, the EEDF is close to Maxwellian in the energy range of a few eV. This is also the range that contributes the most to the integral in (1). In this limit, the electron drift velocity is a monotonically growing function of E/N. One could assume that for NDC to appear, it is necessary for the *e*-*e* collision to compete with the electron-atom energy exchange collision. Figure 1 demonstrates that excited Xe atoms make the NDC effect more pronounced. Their role, because of the relatively low threshold for the process $Xe^* + e \rightarrow Xe^{**} + e$ (1.16 eV), is similar to the role of molecular additives.

The influence of superelastic collisions with metastable atoms on the NDC effect is found to be insignificant as the removal of superelastic collision in our model produces no noticeable effect in our calculations.

One can observe from Fig. 4 that the positions of the minimum in the momentum transfer cross section and the shape of σ_m versus ε curve are quite close for both Xe and Kr. For the case of Ar, the minimum position is lower (0.25 eV against 0.65 eV for Xe) and the rise with respect to energy is not very strong. It is of interest to study how these variations of cross sections affect the existence and amplitude of NDC.

By numerical calculations, the regions in the $\alpha - E/N$ parameter space where NDC exists are located. The calculations are made for Xe (Fig. 5), Kr (Fig. 6), and Ar (Fig. 7). As was shown above, the concentration of the metastable atoms exerts strong influence on the NDC effect. In Figs. 5–7, results for two values of metastable fraction $\delta = \alpha$ and $= 10\alpha$ are illustrated. The populations of higher electronic states are neglected. (It should be noted that the boundaries of specified regions in Figs. 5–7 are drawn by a linear extrapolation procedure between calculated points.)

The role of electron-ion collisions is also studied. It should be noted that in contrast to *e*-*e* collisions, where electron momentum is conserved, the *e*-*i* collisions cause much change of the electron momentum and the effect on electron energy is much less significant. We therefore choose to ignore electron energy change in the *e*-*i* collisions. The momentum transfer cross section for the *e*-*i* collisions in Ar is about 10^{-11} cm^2 at $\varepsilon = 0.25$ eV, which is of the same order as for electron-atom collisions at $\alpha = 10^{-7}$. Calculations show that for Xe and Kr, the role of *e*-*i* collisions may be neglected, and for Ar, these collisions serve to reduce the regions for NDC existence. To illustrate this effect, curve 1 in Fig. 7 is calculated when *e*-*i* collisions are neglected. All other calculations are made by including *e*-*i* collisions.

Let us discuss the data presented in Figs. 5–7 in more detail. Because the results for Xe and Kr are qualitatively similar, we place our attention on the comparison between the NDC effect in Xe and Ar. In the absence of excited atoms, $\delta = 0$, the regions of the NDC existence are oriented along some line that corresponds to the relationship between α and E/N of the type $A(E/N)^s$, where s is approximately 2. The value of s is greater in the case of Xe than in the case of Ar. Additional calculations confirm that this correlation between α and E/N corresponds to the condition that the *e*-*e* collision frequency is of the order of electron relaxation frequency in

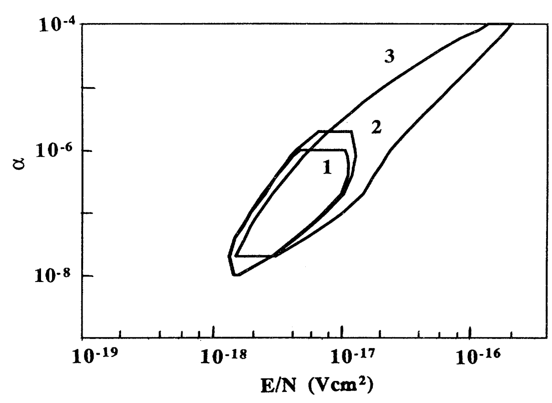

FIG. 6. Region of the NDC effect in $\alpha - E/N$ space for Kr. (1) $\delta = 0$; (2) $\delta = \alpha$; (3) $\delta = 10\alpha$.

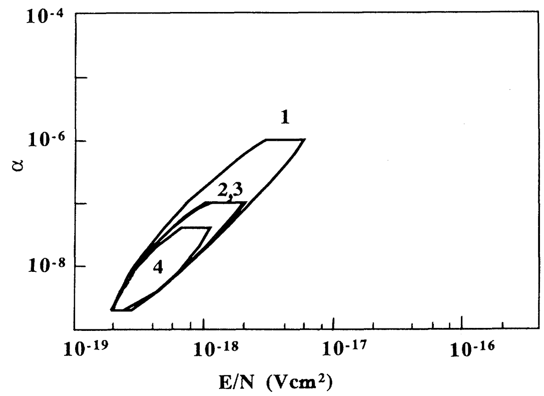

FIG. 7. Region of the NDC effect in $\alpha-E/N$ space for Ar. (1) $\delta=0$, *e-i* collisions neglected; (2) $\delta=0$; (3) $\delta\alpha$; (4) $\delta=10\alpha$.

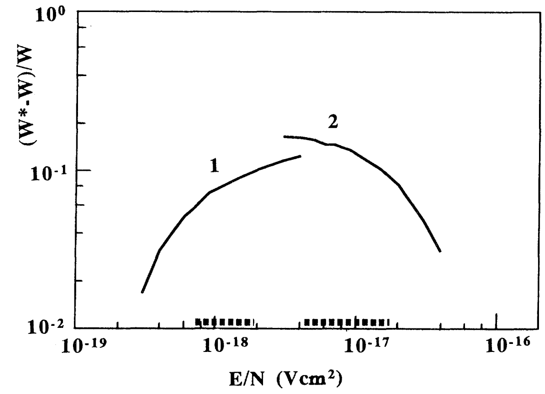

FIG. 8. Reduced w increment induced by metastable atoms $(w^*-w)/w$ as a function of E/N. 1: Ar, $\alpha=8\times10^{-8}$, $\delta=8\times10^{-7}$; 2: Xe, $\alpha=10^{-6}$, $\delta=10^{-5}$. The E/N ranges for the NDC are shown on the E/N axis.

elastic collisions with the main body of the EEDF. The greater the electric field strength, the greater the characteristic electron energy and higher ionization degree is necessary for the *e-e* collisions to compete with the electron-atom collisions.

The difference in the range of E/N where NDC can be observed (compare Figs. 5 and 7) is explained by the difference in positions of the minimum of the momentum transfer cross section (see Fig. 4). The smaller sizes of the NDC region for Ar in comparison with Xe are due to slower increase of the momentum transfer cross section with energy.

Most striking is the deformation of the NDC region with the metastable state populations. It is of opposite signs for Xe and for Ar. The increase of the NDC region with N^* for Xe can be easily explained by the analogy between the electron-excited atom collisions and inelastic collisions with molecular species like N_2 and so on. To understand why the same collisions in Ar give opposite results, it is necessary to look at processes more thoroughly. The momentum transfer cross section for Ar has a much deeper minimum and at a lower energy than for Xe. As a result, the *e-i* collisions come into play at much lower α, and effectively modify the momentum transfer cross section first near the minimum. The cross section for the excitation of the lower metastable state to the higher-lying state has the threshold at 1.5 eV. Calculations predict the existence of the NDC effect in Ar at characteristic electron energies less than 1 eV. It means that for comparable conditions, the role of inelastic collisions with metastable Ar is less important than in the case of Xe. Indeed, at $\delta=\alpha$ in Ar (Fig. 7), there is no difference between curves 2 and 3. At higher excitation degree when $\delta=10\alpha$, the sizes of the NDC region are diminished at the expense of greater E/N and α. In Fig. 8, the increment of w induced by electron-excited atom collisions is shown as function of E/N (the w increment is scaled by w at $d=0$ increments). There is a great difference in these functions for Ar and Xe. In Ar the electron-metastable collisions act as a potential barrier reflecting more effectively electrons with higher energies, which have larger momentum cross sections. As a result, the increment of the electron drift velocity for a stronger electric field is greater than for the lower one. At the same time, the electron-metastable collisions in Xe slow down most of the electrons. Then the increasing momentum transfer cross section causes electron drift velocity to diminish with the electric field.

Let us discuss whether it is possible to observe the predicted NDC in experiments. To estimate chances to observe NDC in a self-sustained discharge, in Fig. 5, the balance between ionization and dissociative recombination processes is plotted as the broken lines for the excitation degree from 0 to 100α. It is supposed that the dominant ion in Xe plasma is Xe_{2^+}. The ionization may occur in two ways: by electron impact from the ground and metastable states. The higher the degree of excitation, the greater the ionization rate because to ionize the metastable, the required energy of the electron is much smaller (3.66 eV) than that for the ionization of the ground state (12.1 eV). It is easy to see that at $\delta=10\alpha$, the ionization-recombination balance condition is compatible with the condition for realizing the NDC effect. Based on this, one can conclude that NDC can in principle be observed in a self-sustained discharge, e.g., in the positive column of a glow discharge at a sufficient large value of pR, where p is the gas pressure and R is the discharge tube radius. However, this conclusion may be violated by the constriction of the positive column, which is associated with the onset of ionization instability when the recombination loss prevails over the diffusion to the walls.

The use of external ionization such as a relativistic electron beam or an x-ray source allows one to control the degree of ionization and electric-field strength independently. In this case, there is no problem realizing the conditions necessary to observe NDC, which should manifest itself as the Gunn-effect instability in the form of discharge current oscillations at characteristic frequencies in the range of 0.1 to 1 MHz [4]. It is well known that in the glow discharge there are regions with abnormally low electric field. These regions are the negative glow and the neighboring dark space, which is a transition zone from the negative glow to the positive column of the discharge. Our estimates show that for gas pressures greater than 1 torr, some region should exist where NDC could take place. It is more probable that this region could be found for discharges in Xe and Kr. The amplitude and frequency of oscillations anticipated depend, among others, on the length of the region with the NDC.

IV. CONCLUSION

In conclusion, the main results of this study are as follows. A mechanism for the NDC formation is proposed for plasmas of pure rare gases (viz., Xe, Kr, or Ar). The amplitude and the necessary conditions for observing the NDC effect are predicted by means of numerical calculations of EEDF, taking into account *e-e*, *e-i*, electron-excited state, and electron-ground-state collisions. The possibility of observing NDC experimentally is discussed briefly.

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