RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 31, NUMBER 3 MARCH 1985

Transient negative mobility of hot electrons in gaseous xenon

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(Received 17 December 1984)

Time-resolved conductivity measurements on nanosecond pulse-ionized xenon gas confirm the prediction of McMahon and Shizgal that initially formed hot electrons can undergo a net displacement in the reverse field direction. This negative mobility situation persists for 30 ns after the pulse at 10 atm ($E/N \lesssim 10^{-19}$ V cm^2) and gradually disappears as the field strength is increased.

INTRODUCTION

Owing mainly to the great advances which have been made in computational capabilities in recent years, there has been an upsurge of theoretical interest in the dynamic aspects of the degradation of the excess energy of initially hot electrons in gaseous systems.[1–8] The problem is basic to a full understanding of the physical and chemical processes occurring in natural and artificial discharges and plasmas and in media subjected to a flux of ionizing radiation. The last aspect is particularly relevant at the present time because of the rapid development and application of subnanosecond pulsed sources of ionizing radiation including electron accelerators, synchroton light sources, and high-intensity lasers.

Because of their apparent simplicity, in the sense that only elastic collisions need to be considered, the rare-gas systems have been taken as the initial proving ground for theoretical treatments. These systems are also appropriate because of the large body of information which is already available from electron beam and swarm studies.[9] Unfortunately, to date, very few direct experimental studies of electron thermalization dynamics have been carried out in sufficient detail to check the predictions of theoretical calculations.

In a recent paper McMahon and Shizgal[10] made a particularly interesting prediction of an effect which should be observed in dynamic thermalization studies for the heavier rare gases. According to these authors, for gases displaying a strong positive power dependence of collision frequency on electron velocity, electrons formed with an initially isotropic velocity distribution should undergo a net displacement in the reverse field direction resulting in effect in a negative mobility.

We report here results of a nanosecond time-resolved pulse radiolysis conductivity study of xenon gas which demonstrate the occurrence of this negative mobility effect.

EXPERIMENT

The experiments were carried out using a parallel plate, two-electrode conductivity cell capable of sustaining high vacuum (10^{-6} Torr) and high pressure (up to 300 atm). The gas between the electrodes was ionized by x rays from a a platinum target subjected to a short pulse (10 or 20 ns) of 3-MeV electrons from a Van de Graaff accelerator. The resulting conductivity transient was amplified using a Keithley 105 amplifier and subsequently monitored using a Tektronix transient digitizer. The overall rise time of the detection system was approximately 5 ns. All data were obtained using single x-ray pulses which resulted in an initial electron concentration of approximately 5×10^9 electrons per cm^3. The xenon was purified before use by passage over calcium and titanium at 600 °C.

RESULTS AND DISCUSSION

In the present work the parameter which is measured is the displacement current resulting from the motion of ions and electrons following pulsed ionization of the medium between two parallel plate electrodes. The displacement current is directly related to the conductivity of the medium which, since electrons are the major charge carrier, is given by

$$\sigma = eN_e\mu_e \ , \tag{1}$$

where e, N_e, and μ_e are the electronic charge and electron concentration and mobility, respectively. The time scale of the measurements is such that electron removal processes due to diffusion, electron-ion recombination, or drawout by the field are negligible. The transient signal may, therefore, be taken to be directly proportional to the mobility of the electrons at a particular time following the pulse. Any change with time of the mobility reflects a change in the average electron atom collision frequency ν_m as the excess energy of the electrons is degraded according to

$$\mu_e = \frac{2}{3}\frac{e}{m}\frac{1}{\langle \nu_m \rangle} \ , \tag{2}$$

with $\langle \nu_m \rangle$ given by

$$\frac{1}{\langle \nu_m \rangle} = \int_0^\infty \frac{1}{\nu_m(u)} u^{3/2} \frac{df(u)}{du} du \ , \tag{3}$$

where $f(u)$ is the electron-energy-distribution function.

In Fig. 1 are shown the mobility transients observed following pulsed ionization of xenon at 10.0 atm with the applied field varying from 13.3 to 106.7 V/cm corresponding to density reduced field strengths, E/N, of 5.14×10^{-20} and 41.2×10^{-20} V cm^2. The transients are characterized by a rapid increase following the pulse to a maximum value at 70–100 ns and a subsequent decay to an eventual constant level.

As in the case of argon, which has been previously reported,[11] the behavior observed is attributed to the formation of

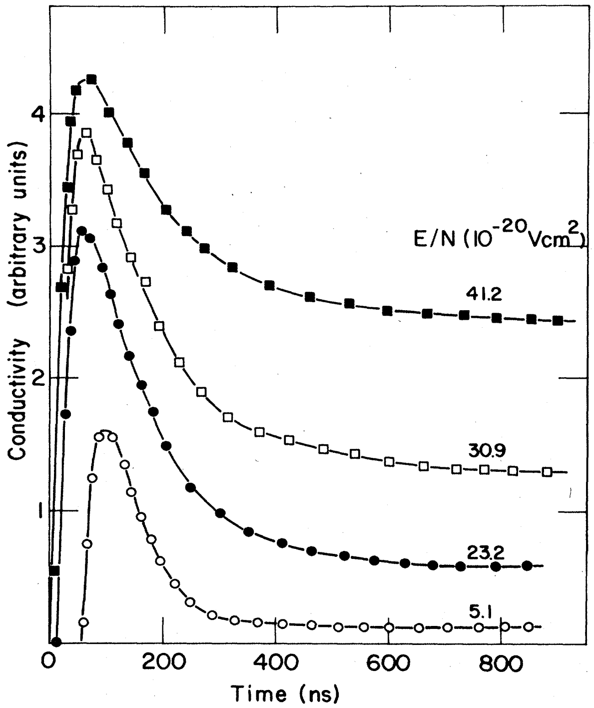

FIG. 1. Conductivity transients in pulse ionized xenon at 10 atm and 295 K for the density-reduced field strengths shown. Zero time corresponds to the beginning of the 20-ns x-ray pulse.

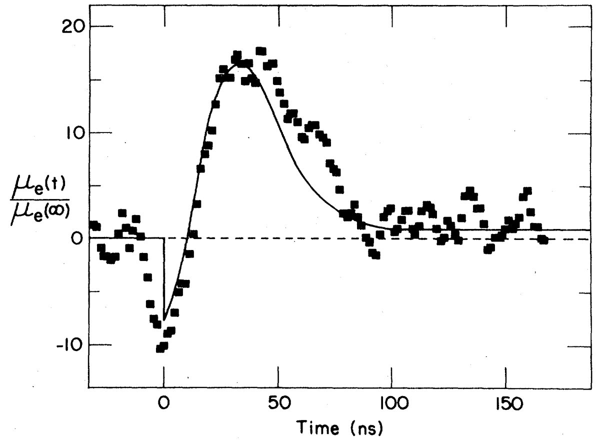

FIG. 2. Full line: the calculated time dependence of the "zero field" electron mobility relative to the eventual thermal mobility according to McMahon and Shizgal, Ref. 10, for $N_{Xe} = 5.74\times10^{20}$ cm^{-3}. Electrons are taken to be formed in a δ pulse with an initial energy of 0.864 eV and a δ-function distribution of velocities. Points: experimental data of the time dependence of the conductivity of xenon (5.74×10^{20} cm^{-3}) following ionization with a 10-ns pulse of x rays (time is from end of pulse) for $E/N = 1.16\times10^{-20}$ V cm^2. The data have been normalized vertically to give roughly the same height at maximum signal as for the calculated curve.

subexcitation electrons with an initial average energy considerably higher than the energy at which the Ramsauer minimum in the electron-atom collision frequency occurs[9] (ca. 0.6 eV for xenon). The average collision frequency, therefore, passes through a minimum (mobility through a maximum) as the average electron energy cools down from super-Ramsauer to sub-Ramsauer energies. The eventual, constant mobility corresponds to the average collision frequency determined by the static, equilibrium electron-energy distribution prevailing at a given applied field.

Two aspects of the results are of particular interest. Firstly, the significant increase in the time required to reach the equilibrium mobility with increasing field strength, and, secondly, the apparent delay before the initial growth occurs, which is evident at low field strengths. This latter effect which appears as a delay because of the automatic subtraction of the baseline from the experimental traces was found to be due to a large initial negative going signal. This effect was absent in helium and argon and could not be attributed to an artifact of the experiment. It remained, therefore, as a curiosity until the recent theoretical calculations of McMahon and Shizgal.[10]

In their work the latter authors calculated the expected dynamic behavior of the electron mobility based on measured collision cross-section data for several gases. For those atoms displaying a pronounced Ramsauer minimum they found, for the initial conditions of an isotropic velocity distribution with a corresponding energy in excess of the Ramsauer energy, that the mobility of the electrons could have a transient negative value. This can be understood in terms of Eq. (3) which can lead to negative values of $\langle \nu_m \rangle$ if the power dependence of $\nu_m(u)$ on energy is sufficiently positive. For example, for a Maxwellian distribution this would occur for a dependence of the form $\nu_m(u) = au^n$ with $n > \frac{5}{2}$. The extremely rapid rise of the collision frequency above the Ramsauer minimum in the heavier rare gases is, therefore, the source of negative contributions to the mobility for electrons in this energy regime.

McMahon and Shizgal have carried out quantitative calculations of $\mu_e(t)/\mu_e^{th}$ for gaseous xenon under conditions of "zero field" and these values are given as the solid line in Fig. 2. In order to compare the calculated values with experiment we have chosen the data which we have available for the lowest value of the density reduced field strength at which measurements have been made. This was for a xenon pressure of 20.0 atm and a field of 6.67 V cm^{-1} giving $E/N = 1.16\times10^{-20}$ V cm^2. The experimental data have been normalized to give the same maximum value of the transient as in the calculations.

As can be seen from Fig. 2 the form of the experimental transient is very similar to that predicted, including the extent and the duration of the negative mobility transient. On inspection of the dependence of the shape of the calculated curve on the value taken for the initial electron energy, as given by McMahon and Shizgal, it is apparent that an even better fit to experiment would be obtained if a somewhat higher initial energy were taken than the highest value of approximately 0.9 eV used to derive the curve in Fig. 2. It is worth emphasizing that the time scale in Fig. 2 is absolute and is identical for both the theoretical and experimental results. Extension of the theoretical treatment to the case of nonzero field would be of considerable interest.

In conclusion, it can be said that the phenomenon of a negative electronic mobility, while conceptually somewhat difficult, is practically realizable although it is of essense transitory in nature.

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