論 文
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라디오 주파수전계에 의한 질소가스의 브레이크 다운 현상

The Breakdown Phenomena of N_2 gas by RF Electric Field

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요 약

주파수가 13.56MHz인 고주파 전계에 의한 질소 가스의 방전개시현상은 직류전계일때와 매우 다르다. 본 논문은 전자의 분포함수와 확산 방정식을 이용하여 이 현상을 고찰하였다.

볼쯔만 방정식으로 부터 유도된 2계 미분방정식을 풀어서 전자분포함수를 구하고, Kinetic theory를 이용하여 전자의 이온화계수와 확산계수를 구한다. 이온화에 의해서 생성된 전자의 수와 확산에 의해 소멸되는 전자의 수가 같아질때 방전은 일어나므로 전자의 분포함수로부터 구한 이온화계수와 확산계수를 이용하여 압력변화에 따른 방전개시전압을 구할 수 있다.

컴퓨터를 이용하여 이론치를 구한 다음 실험을 통해 이것을 검증하였다.

Abstract

The breakdown phenomena of N_2 gas by 13.56MHz electric field are very different from those under steady field.

In this paper we analyzed the breakdown phenomena by using electron distribution function and diffusion equation. The second-order differential equation derived from the Boltzmann equation is solved for the electron distribution function. The ionization rate and diffusion coefficient are calculated using kinetic theory formulas. The breakdown condition is that the number of electrons produced by ionization equal the number diffusing to the walls of the discharge chamber.

Theses theoretical breakdown electric fields are calculated by the computer and compared with the experimental values.

1. Introduction

In this paper the breakdown phenomena of N_2 gas at 13.56 MHz are analyzed. The ionization processes at 13.56 MHz become very different from those under steady fields. As in the case of a steady field, the type of gas or gas mixture and its total pressure play an important role in the sequence of events that occur.

Since the breakdown at 13.56 MHz in our system--radius of electrode, R, is 4 cm and gap spacing, d is 3.5 cm--can be considered as the diffusion controlled breakdown, the criterion for the breakdown of N_2 gas

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at 13.56 MHz is a condition that the number of ionizations by the collision between electrons and neutral gas molecules equal to the number of the lost electrons by diffusion.

The ionization rate and diffusion coefficient are deduced using the distribution functions which is derived from the Boltzmann equation and are combined with the diffusion equation to predict the breakdown electric fields. These theoretical breakdown electric fields are calculated by the computer and compared with the experimental values.

2. Validity of Diffusion Theory in Our System

Breakdown is classified as mobility-controlled breakdown, diffusion-controlled breakdown, attachment-controlled breakdown, and recombination-controlled breakdown according to the loss mechanism of electron.

When an electric field with a frequency which is above the cut-off frequency is applied to a discharge chamber, electrons will disappear mainly by the diffusion to the wall of the discharge chamber. In this case, because there is no loss of of electrons by the drift to the electrodes, we need not consider the secondary emission of electrons from the electrodes. And electrons will oscillate between the electrodes. According to the Townsend theory, a breakdown occurs when the number of electrons made by the ionization of neutral gas molecules collided by the electrons equals to the number of electrons lost by the diffusion to the wall of discharge.

In our system, cut-off frequency [1] is found as follows;

$$f_{co}=\frac{K_e E_o}{\pi d} \qquad K_e=\frac{e/m}{\nu_m+j\omega} \tag{1}$$

where, K_e is electron mobility and ν_m is momentum collision frequency, ω is the frequency of the electric field. Gap spacing, d, is 3.5 cm and E_0 is about 40 V/cm.

Collision frequency is equal to $N\sigma v$. Here N is the density of N_2 gas and σ is collision cross section and v is velocity. Using the data of the cross section of N_2 gas [2], we find that ν_m is about 5×10^9P, where P is pressure of N_2 gas in Torr. f_{co} becomes 1-10 MHz in the range of N_2 gas pressure 0.01-1 Torr and it is less than the frequency of our R.F. generator, 13.56 MHz. Thus the breakdown of our system can be considered as diffusion controlled breakdown.

3. Boltzmann Equation

In the high frequency, the motion of ions is insignificant in comparision with that of the electrons because electron's acceleration is normally thousands of times as great as ion's accleleration. Therefore almost all the electrical properties of an ionized gas at high frequencies are determined by the electrons. The electron distribution function, F (**v**, **r**, t), is defined as the number of electrons in a volume element of space, dxdydz, having velocities between **v** and **v**+d**v**. If F is not a function of the space variables, the distribution is uniform and if it is a function of speed and not of velocity, it is isotropic.

In general the mean value of any quantity, Q (**r**, t), is given by an integral involving the distribution function;

$$n(\mathbf{r},\ t)\ \langle Q(\mathbf{r},\ t)\rangle=\int F(\mathbf{r},\ \mathbf{v},\ t)Q(\mathbf{r},\ t)d\mathbf{v}$$

where, n(**r**, t) represents density.

The exact form of the electron distribution function can be derived through consideration of the gain and loss of electrons from an incremental volume in phase space defined by d^3rd^3v. The equation governing the net transfer of electrons from this volume is known as the Boltzmann equation and is expressed as [3];

$$C=\frac{\partial F}{\partial t}+\mathbf{v}\cdot\nabla_r F+\mathbf{a}\cdot\nabla_v F \tag{2}$$

where ∇_r is a gradient in x, y, z space and ∇_v is a gradient in velocity space and a is the acceleration, C is the time rate of change of F by collisions.

The Boltzmann equation must be solved in order to obtain the electron distribution function. Since no exact solution to this equation is known, an approximate solution must be sought. A technique which has been used extensively considers that the solution can be expressed as the sum of an isotropic portion plus a small anisotropic perturbation [4]. Under such a consideration it is logical to expand the distribution function in spherical harmonics. The first two terms in such an

expression are sufficient to give an accurate representation of F and lead to a satisfactory explanation of the electrical phenomena in the plasma [2].

$$\mathbf{F}=\sum_{nm}\sum \mathbf{F}_{n,m}(\mathbf{r},\ t)Y_n^m(\theta,\ \varphi) \tag{3}$$

where $Y_n^m(\theta,\ \varphi)=(-1)^m\sqrt{\frac{2n+1}{4\pi}\frac{(n-m)!}{(n+m)!}}P_n^m(\cos\theta)e^{jm\varphi}$

and $P_n^m(\cos\theta)$ are the associated Legendre polynomials. The first few spherical harmonics in terms of velocity components are;

$$Y_0^0=1$$

$$Y_1^0=\frac{v_z}{v},\ Y_1^1=\frac{v_x}{v},\ Y_1^{-1}=\frac{v_y}{v}$$

$$Y_2^0=\frac{2v_z^2-v_x^2-v_y^2}{2v^2}$$

$$Y_2^1=\frac{3v_xv_z}{v^2},\ Y_2^2=3\frac{v_x^2-v_y^2}{v^2}$$

$$Y_2^{-1}=3\frac{v_yv_z}{v^2},\ Y_2^{-2}=6\frac{v_xv_y}{v^2}$$

We can express F in the following way;

$$\mathbf{F}=\mathbf{F}_{0,0}+\frac{v_x}{v}\mathbf{F}_{1,x}+\frac{v_y}{v}\mathbf{F}_{1,y}+\frac{v_z}{v}\mathbf{F}_{1,z}+\cdots$$
$$=\mathbf{F}_0+\frac{\mathbf{v}\cdot\mathbf{F}_1}{v}+\cdots \tag{4}$$

The term $\mathbf{F}_0$ represents the isotropic part and the terms involving $\mathbf{F}_1$ express the small perturbations.

In a similar way, we now make an expression for the collision terms.

$$C=C_0+\frac{\mathbf{v}\cdot\mathbf{C}_1}{v}+\cdots \tag{5}$$

By substituting (4) and (5) into (2), we can rewrite Boltzmann equation involving zero and first order terms as follows;

$$C_0+\frac{\mathbf{v}\cdot\mathbf{C}_1}{v}=\frac{\partial\mathbf{F}_0}{\partial t}+\frac{v}{3}\nabla\cdot\mathbf{F}_1+\frac{e}{3mv^2}\frac{\partial}{\partial v}(v^2\mathbf{E}\cdot\mathbf{F}_1)$$
$$+\frac{\mathbf{v}}{v}\cdot\left(\frac{\partial\mathbf{F}_1}{\partial t}+v\nabla\mathbf{F}_0+\frac{e}{m}\mathbf{E}\frac{\partial\mathbf{F}_0}{\partial v}\right) \tag{6}$$

The electric field, $\mathbf{E}=\mathbf{E}_p\ \exp(j\omega t)$, introduces another variation. The phenomena which result from the interaction of an alternating field and charged particles occur at frequencies $n\omega/2\pi$, where n may be 0, 1, 2, ... We therefore make a Fourier expansion in time for each spatial component.

$$\mathbf{F}_n=\mathbf{F}_n^0+\mathbf{F}_n^1e^{j\omega t}+\mathbf{F}_n^2e^{2j\omega t}+\cdots \tag{7}$$

With introduction of a Fourier expansion into Boltzmann equation involving the spherical harmonics, we can obtain the following set of coupled Eqs.;

$$C_0^0=\frac{\partial\mathbf{F}_0^0}{\partial t}+\frac{\mathbf{v}}{3}\nabla\cdot\mathbf{F}_1^0+\frac{e}{6mv^2}\frac{\partial}{\partial v}(v^2\mathbf{E}_p\cdot\mathbf{F}_1^1) \tag{8-1}$$

$$C_0^1=\frac{\partial\mathbf{F}_0^1}{\partial t}+j\omega\mathbf{F}_0^1+\frac{\mathbf{v}}{3}\nabla\cdot\mathbf{F}_1^1+\frac{e}{3mv^2}\frac{\partial}{\partial v}(v^2\mathbf{E}_p\cdot\mathbf{F}_1^0) \tag{8-2}$$

$$\mathbf{C}_1^0=\frac{\partial\mathbf{F}_1^0}{\partial t}+v\left(\nabla\mathbf{F}_0^0+\frac{e}{2m}\mathbf{E}_p\frac{\partial\mathbf{F}_0^1}{\partial v}\right) \tag{8-3}$$

$$\mathbf{C}_1^1=\frac{\partial\mathbf{F}_1^1}{\partial t}+j\omega\mathbf{F}_1^1+v\nabla\mathbf{F}_0^1+\frac{e}{m}\mathbf{E}_p\frac{\partial\mathbf{F}_0^0}{\partial v} \tag{8-4}$$

These four equations can be simplfied by introducing a new variable $u=mv^2/2e$ (volts). Then we have

$$C_0^0=\frac{\partial\mathbf{F}_0^0}{\partial t}+\frac{v}{3}\left(\nabla\cdot\mathbf{F}_1^0+\frac{1}{2u}\frac{\partial}{\partial u}(u\mathbf{E}_p\cdot\mathbf{F}_1^1)\right) \tag{9-1}$$

$$C_0^1=\frac{\partial\mathbf{F}_0^1}{\partial t}+j\omega\mathbf{F}_0^1+\frac{v}{3}\left(\nabla\cdot\mathbf{F}_1^1+\frac{1}{u}\frac{\partial}{\partial u}(u\mathbf{E}_p\cdot\mathbf{F}_1^0)\right) \tag{9-2}$$

$$\mathbf{C}_1^0=\frac{\partial\mathbf{F}_1^0}{\partial t}+v\left(\nabla\mathbf{F}_0^0+\frac{1}{2}\mathbf{E}_p\frac{\partial\mathbf{F}_0^1}{\partial u}\right) \tag{9-3}$$

$$\mathbf{C}_1^1=\frac{\partial\mathbf{F}_1^1}{\partial t}+j\omega\mathbf{F}_1^1+v\left(\nabla\mathbf{F}_0^1+\mathbf{E}_p\frac{\partial\mathbf{F}_0^0}{\partial u}\right) \tag{9-4}$$

4. Collision Integral

Since the collision integral plays a very important role in the determination of the form of the distribution function, we must determine its explicit form and we first consider elastic collision. Massey and Burshop [2] have calculated the collision integral C_0 and C_1 for elastic collisions. They considered two contributions; the number of particles/s scattered out of $d^3\mathbf{v}$ at $d^3\mathbf{r}$ by collisions (a loss term) and the number of particles/s scattered into $d^3\mathbf{v}$ at $d^3\mathbf{r}$ by collision (5) (6). Their analysis lead to

$$C_{0,el}=\frac{m}{M}\frac{2}{u^{\frac{1}{2}}}\frac{\partial}{\partial u}(u^{\frac{3}{2}}\nu_m F_0)$$

where m and M are the masses of the electron and molecule, respectively.

The vector component is given more simply by

$$\mathbf{C}_{1,el}=-\nu_m\mathbf{F}_1$$

The effects of inelastic collisions may be represented by

$$C_{o,in} = -h\nu_c F_0$$

where h is the sum of the efficiencies of the inelastic collision processes and ν_c is collision frequency.

5. Differential Equation for Distribution Function

Introduction of the results of collision integral into the set of differential equation (9) results in a coupled set of differential equations. The term F_0^1 represents the first harmonics of the spherically symmetrical part of the distribution function. In the absence of a dc field there are no physical processes in the discharge which this term would represent [7]. Therefore we may neglect F_0^1.

Breakdown problems can generally be solved using steady state distribution function, so we may set the time derivative terms equal to zero. When we make these simplifications, equation (9) reduced to the following.

$$\frac{m}{M}\frac{2}{u^{\frac{1}{2}}}\frac{\partial}{\partial u}(u^{\frac{3}{2}}\nu_m F_0^0) - h\nu_c F_0^0 = \frac{v}{3}\left(\nabla\cdot\mathbf{F}_1^0 + \frac{1}{2u}\frac{\partial}{\partial u}(u\mathbf{E}_p\cdot\mathbf{F}_1^1)\right) \quad (10\text{-}1)$$

$$\nabla\cdot\mathbf{F}_1^1 = -\frac{1}{u}\frac{\partial}{\partial u}(u\mathbf{E}_p\mathbf{F}_1^0) \quad (10\text{-}2)$$

$$-\nu_m\mathbf{F}_1^0 = v\nabla F_0^0 \quad (10\text{-}3)$$

$$-\nu_m\mathbf{F}_1^1 = j\omega\mathbf{F}_1^1 + v\mathbf{E}_p\frac{\partial F_0^0}{\partial u} \quad (10\text{-}4)$$

Substituting Eqs. (10-2) to (10-4) into (10-1), we have

$$\frac{v}{3}\left(\frac{E_p^2}{2u}\frac{\partial}{\partial u}\left(uv\frac{m}{\nu_m^2+\omega^2}\frac{\partial F_0^0}{\partial u}\right) + \frac{v}{\nu_m}\nabla^2 F_0^0\right) = h\nu_c F_0^0 - \frac{2m}{M}\frac{1}{u^{\frac{1}{2}}}\frac{\partial}{\partial u}(u^{\frac{3}{2}}\nu_m F_0^0) \quad (11)$$

Since the spatial variation may be separated out, we may replace $F(u,x,y,z) = f(u)g(x,y,z)$. With this change, we have a second order differential equation for f, the energy varying component of the spherical term in the distribution.

$$\frac{2e}{3m}\frac{E^2}{u^{\frac{1}{2}}}\frac{d}{du}\left(\frac{u^{\frac{3}{2}}\nu_m}{\nu_m^2+\omega^2}\frac{df}{du}\right) + \frac{2m}{M u^{\frac{1}{2}}}\frac{d}{du}(u^{\frac{3}{2}}\nu_m f) = \left(h\nu_c + \frac{2eu}{3m\nu_m\wedge^2}\right) \quad (12)$$

In order to solve equation (12), we must know the expression of the collision frequencies. The collision frequency is related to the collision cross section as $\nu = N\sigma v$, where N is the density of neutral atom at 1 Torr pressure. The cross section $\sigma(m^2)$ may to found from the collision probability P_c by multiplying $2.82 + 10^{-21}$, provided that the mean free path is specified in meters and the pressure in Torr. The collision probability is defined by setting $1P_c p = 1$, where 1 is the mean free path and p is the pressure.

The variation of the collision probability P_c for electrons with molecules of nitrogen as a function of electron energy is shown in figure 1 [2]. As shown in figure 2., we modified the data of Brode in the straight lines for a convenience in calculation. Using these data of the collision probability, we may express the momentum transfer collision frequency as follows;

a. $u < 2.6$ eV

$$\nu_m = 5.519\times10^7 p u^{\frac{1}{2}}(35.88u - 4.29) \quad (13\text{-}1)$$

b. $2.6 < u < 5.2$ eV

$$\nu_m = 5.519\times10^7 p u^{\frac{1}{2}}(-21.54u + 145) \quad (13\text{-}2)$$

c. $5.2 < u < 25$ eV

$$\nu_m = 5.519\times10^7 p u^{\frac{1}{2}}(0.5u + 30.37) \quad (13\text{-}3)$$

d. $25 < u < 36$ eV

$$\nu_m = 5.519\times10^7 p u^{\frac{1}{2}}(-0.45u + 54.36) \quad (13\text{-}4)$$

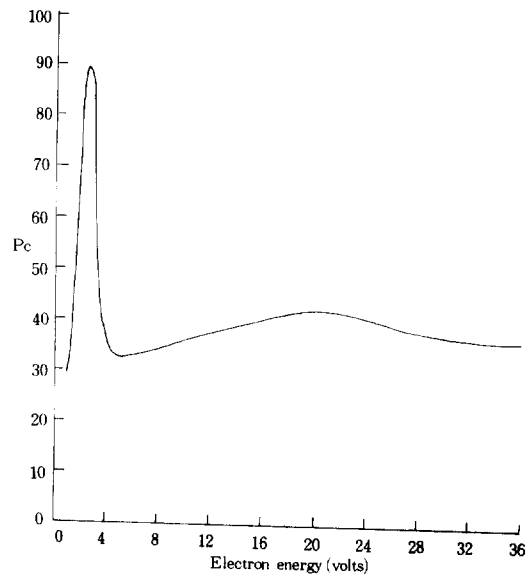

Fig. 1. Probability of collision for electrons in nitrogen.

Substitution of the above value of collision frequencies into equation (12) leads to the following Eqs. .

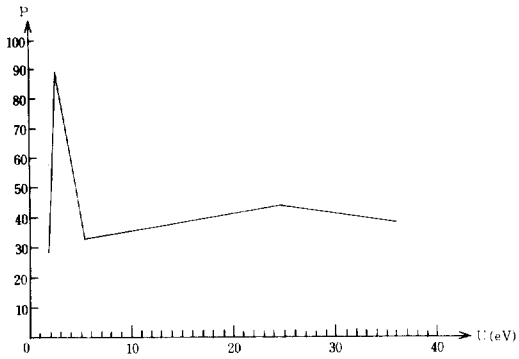

Fig. 2. Probability of collision for electrons in N_2.

a) $u<2.6eV$

$$\frac{d^2f}{du^2}+\left(\frac{3}{2u}+\frac{(53.82u-2.15)(7.26-3.05p^2u(35.9u-4.29)^2)}{u(35.9u-4.29)(3.04p^2u(35.9u-4.29)^2+7.26)}+\frac{0.33}{E^2}(3.05p^2u(35.9u-4.29)^2+7.26)\right)\frac{df}{du}+\frac{3.05p^2u(35.9u-4.29)^2+7.26}{E^2}\left(\frac{35.63u-2.84}{u(35.9u-4.29)}-\frac{14577.3}{3.05p^2u(35.9u-4.29)^2}\right)f=0 \quad (14\text{-}1)$$

b) $2.6<u<5.2eV$

$$\frac{d^2f}{du^2}+\left(\frac{3}{2u}+\frac{(-32.3u+72.5)(7.26-3.05p^2u(-21.54u+145)^2)}{u(-21.54+145)(3.05p^2u(-21.54u+145)^2+7.26)}+\frac{0.33}{E^2}(3.05p^2u(-21.54u+145)^2+7.26)\right)\frac{df}{du}+\frac{(3.05p^2u(-21.54u+145)^2+7.26)}{E^2}\left(\frac{-15.43u+96}{u(-21.54u+145)}-\frac{14577.3}{3.05p^2u(-21.54u+145)^2}\right)f=0 \quad (14\text{-}2)$$

c) $5.2<u<25eV$

$$\frac{d^2f}{du^2}+\left(\frac{3}{2u}+\frac{(0.76u+15.18)(7.26-3p^2u(0.5u+30.37)^2)}{u(0.5u+30.37)(3p^2u(0.5+30.37)^2+7.26)}+\frac{0.33}{E^2}(3.05p^2u(0.5u+30.37)^2+7.26)\right)\frac{df}{du}+\frac{(3.05p^2u(0.5u+30.37)^2+7.26)}{E^2}\left(\frac{0.5u+20}{u(0.5u+30.37)}-\frac{14577.3}{3.05p^2u(0.5u+30.37)^2}\right)f=0 \quad (14\text{-}3)$$

d) $25<u<36eV$

$$\frac{d^2f}{du^2}+\left(\frac{3}{2u}+\frac{(-0.68u+27.18)(7.26-3.05(p^2u+54.36)^2)}{u(-0.45u+54.36)(3.05p^2u(-0.54u+54.36)^2+7.26)}+\frac{0.33}{E^2}(3.05p^2u(-0.45u+54.36)^2+7.26)\right)\frac{df}{du}+\frac{(3.05p^2u(-0.45u+54.36)^2+7.26}{E^2}\left(\frac{-0.45u+36}{u(-0.45u+54.36)}-\frac{14577.3}{3.05p^2u(-0.45u+54.36)^2}\right)f=0 \quad (14\text{-}4)$$

6. Breakdown Criterion of N_2 gas

The breakdown condition is that the number of electrons produced by ionization equals the number of electrons diffusing to the walls of the container. This breakdown condition is combined with a solution of the diffusion equation, the ionization rate and diffusion coefficient to obtain an equation which results in the breakdown electric field.

The ionization rate, ν_i, and diffusion coefficient, D, are calculated using standard kinetic theory formulas to yield the following equation.

$$n\nu_i=\int_0^\infty h_i\nu_c f4\pi v^2dv=\frac{2\pi}{3}\left(\frac{2e}{m}\right)^{\frac{5}{2}}E^2\left(\frac{u^{\frac{3}{2}}\nu_m}{\nu_m^2+\omega^2}\frac{df}{du}\right)_{u=u_i} \quad (15\text{-}1)$$

$$nD=\int_0^\infty (Lv/3)f4\pi v^2dv=\frac{2\pi}{3}\left(\frac{2e}{m}\right)^{\frac{5}{2}}\int_0^{u_i}\frac{u^{\frac{3}{2}}f}{\nu_m}du \quad (15\text{-}2)$$

where h_i : efficiency of ionization
E : rms of electric field
L : mean free path

The breakdown of the gas will materialize when the production of newly ionized particles barely exceeds or is just equal to the rate of their loss by all deionizing processes, including diffusion. Deionization can also occur by recombination and attachment. When these two mechanism can be neglected in our system, deffusion becomes the only loss factor. For a cylindrical geometry, we may write down a condition for breakdown using Eq.(15) as follows;

$$\frac{\nu_i}{D}=\frac{E^2\left(\frac{u^{\frac{2}{3}}\nu_m}{\nu_m^2+\omega^2}\frac{df}{du}\right)_{u=u_i}}{\int_0^{u_i}\frac{u^{\frac{3}{2}}f}{\nu_m}du}=\frac{1}{\Lambda^2} \quad (16)$$

where Λ is the characteristic diffusion length. For a cylindrical geometry, the characteristic diffusion lenght is espressed as follows[1];

$$\frac{1}{\Lambda^2}=\left(\frac{2.405}{R}\right)^2+\left(\frac{\pi}{d}\right)^2$$

R ; radius of electrode
d ; gap spacing

7. Theoretical and Experimental Result

The schematic presentation of our experiment is shown in figure 3, in which the upper electrode is powered and the lower one is grounded.

An RF power of 13.56MHz is applied to the electrodes through the L-matching network and the Langmuir probe is used to measure the plasma parameters.

Under the assumption that the electric field is uniform between the electrodes, we obtained the breakdown electric field by measuring the breakdown voltage with an oscilloscope.

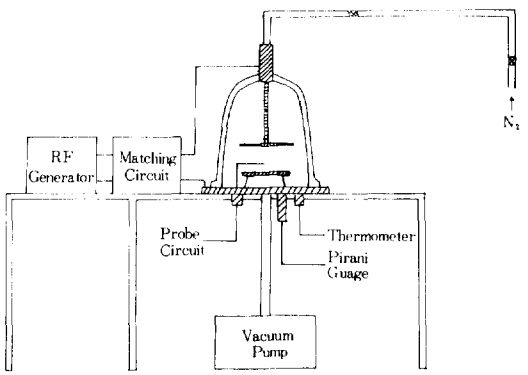

Fig. 3. Discharge system.

Figure 4. shows the result of theory and experiment of breakdown electric fields as a function of pressure. We found the result of theory by solving Eq. (12) and (13) through the numerical analysis using the Runge-Kutta method.

As shown in figure 4., the electric fields have a minimum value at a point where pressure is about 0.1 Torr. As pressure exceeds this value, electric field increases because the mean free path decreases and electrons can not gain sufficient energy from fields. As pressure decreases, electric field also increases because electrons have a smaller chance for collision with neutral molecules. In spite of small error, the agreement between theory and experiment in figure 4. verifies the correctness of the approach.

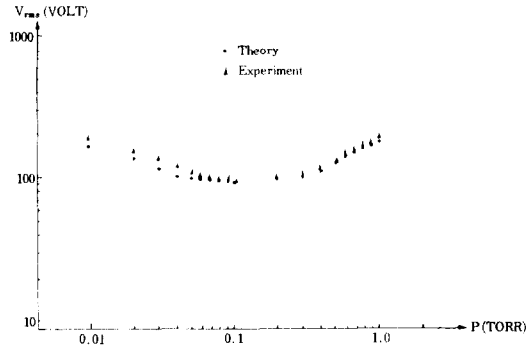

Fig. 4. Theory and experiment at 13.56MHz.

8. Conclusion

The breakdown electric fields of N_2 gas by 13.56 MHz electric field has been calculated theoretically on the basis of the electron continuity equation, considering diffusion to the discharge walls as the only removal process. This approach, containing no adjustable nitrogen discharge data or constant, and involving only the ionization energy and collision cross section, enables us to predict breakdown electric fields. It has been derived from the electron distribution function given by kinetic theory, and the diffusion equation.

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