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A STUDY OF THE ELECTRICAL CONDUCTIVITY IN HEAVY NOBLE GAS PLASMAS

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Abstract: A comparative experimental and theoretical study of the electrical conductivity of a high density noble gas plasma is reported. Theoretical calculations were carried out using available theories and it was found that systematic discrepancies between theory and experiment still exist.

1. Introduction

Heavy noble gas plasmas have been the subject of detailed investigations of many authors[1,2]. However, some problems concerning transport phenomena still remain unsolved. For example, there are systematic discrepancies between experimental values of electrical conductivity and those calculated on the basis of kinetic theory, at electron densities above 10^{16} cm^{-3}. This fact was observed on cascade arcs as well as on pulsed arcs[3-6], but has not yet obtained a satisfactory explanation.

In low temperature plasmas of such high densities the condition $n_D \gg 1$ (where n_D is the number of particles in a sphere of the Debye radius) is not fulfilled[7]. This means that the Coulomb field of a charged particle is not screened at the distance $r = r_D$ (r_D is the Debye radius), so that the interaction potential can not be expressed in a Debye-Hückel form

$$\varphi(r) = \frac{e_1 e_2}{r} \exp\left(-\frac{r}{r_D}\right), \tag{1}$$

as in a low density (ideal) plasma. In fact, a correct form of the interaction potential in this case is still unknown. Some attempts have been made to express this potential in a form taking into account the screening, where, instead of r_D, one introduces a screening radius optimized by diminishing the discrepancy between experimental evidence and theory of electrical conductivity[5,7]. With the interaction potential obtained in such a way, all other transport properties of plasma can be calculated. To do this, one has to eliminate all other calculation difficulties in connection with high density effects (proper form of collision integral, changes in plasma equilibrium composition etc.).

On the other hand, considerable effort has been made to give a theoretical explanation of phenomena occurring in plasmas at high densitities. Important corrections of classical kinetic theory concerning nonideality effects were obtained by several authors. Klimontovich[8] formulated generalized Boltzmann and Landau equations with pair-collision approximation, by taking into account the particle correlation in the interval between two consecutive collisions (not only at the moment of the collision). In a more recent paper[9] the same author proposed a model collision integral for charged particle collisions, where the screening of electrons by heavy, slow ions was neglected. A similar assumption has recently been made by Devoto[6], who also suggested that the screening by ions should be neglected. Kaklyugin and Norman[10] found that the space correlation between colliding particles results in the localization of a fraction of electrons and hence their elimination from the electric energy transport. All these effects have certainly some influence on the electrical conductivity, and it is the main purpose of this paper to estimate this influence in heavy noble gases plasmas. Both experimental and theoretical aspects of the problem have been analyzed and the results obtained are given in the form of temperature dependence of electrical conductivity.

2. *Theoretical considerations*

Ideal plasma. For the calculations of the electrical conductivity use was made of the Shkarofsky method[11]. Basic calculations were carried out for an ideal plasma and the nonideality effects were introduced through changes in collision integrals, the equilibrium composition and distribution functions. The starting equation was the Boltzmann kinetic equation, where electron-atom collisions were represented by the Boltzmann collision integral, while collisions between charged particles were represented by a Focker-Planck collision term. The standard procedure[11,12] of expanding the distribution function

$$f = f_0 + \frac{\vec{v}}{v}\vec{f}_1 + \frac{\vec{v}\,\vec{v}}{v^2} : \hat{f}_2 + \dots \tag{2}$$

was used, where the equation was solved for f_1 using the following representation of f_1

$$f_1 = w^{1/2} \frac{f_0(w)}{\beta} \sum_r q_r L_r^{3/2}(w) \; ; \; w = \frac{m v^2}{2 \varkappa T} = \beta v^2, \tag{3}$$

where $L_r^{3/2}$ are Laguerre-Sonine polynomials of the order 3/2, and q_r are coefficients to be determined. Electron-ion and electron-electron collisions were introduced in the same way as in Ref.[12], while electron-atom collisions have been introduced previously only for the case where momentum-transfer cross-sections were of the form[11] $Q_m \sim v^n$, or where they could be represented by a polynomial in energy[13]. However, both assumptions are valid only in small energy intervals, especially for gases which exhibit a pronounced Ramsauer effect.

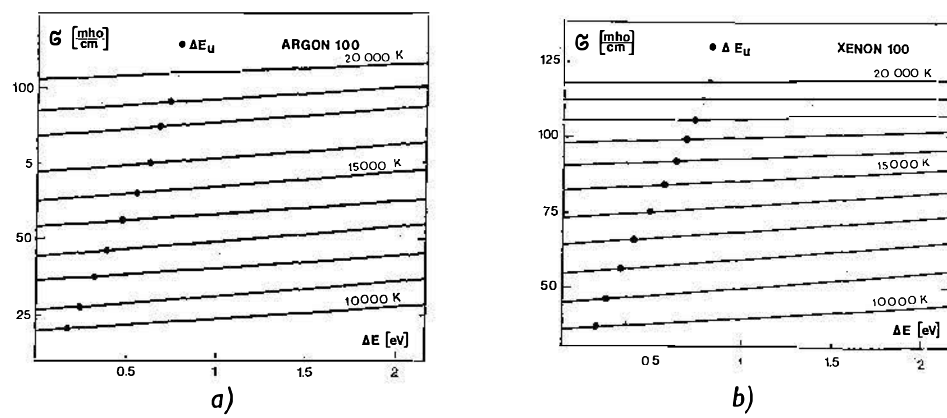


Fig. 1. Variation of the calculated electrical conductivity with lowering ionization potential for different temperatures. Initial pressure 100 torr. Solid circles represent the values of ΔE from formula (6) for given temperature; a) Argon, b) Xenon.

The problem is that momentum-transfer cross-sections for electron-atom collisions are not quite well established. Theoretical expressions are available only for the low energy region[14]; besides, experimental data for Ar, Kr, and Xe show considerable discrepancies, and it is very difficult to decide which are the most accurate. Hence numerical integration of particular experimental data is not very suitable, and we preferred to find out some empirical formula. Considering various experimental and theoretical data[14–18], we found that the best fit to these data may be obtained by the formula

$$Q_m = a \exp(-bE) + cE^d \exp(-fE), \tag{4}$$

where Q_m is in Å^2, and E in eV. The numerical values of the coefficients are given in the Table.

Table.

	a	b	c	d	f
Ar	8.05	28.024	0.99	1.743	0.136
Kr	30.7	11.6	0.79	2.06	0.15
Xe	108.41	14.5	3.456	1.83	0.191

Using the relation

$$L_r^{3/2}(w)\, L_s^{3/2}(w) = \sum_k (-1)^k \beta_k^{(r,s)} w^k,$$

where $\beta_k^{(r,s)}$ are the coefficients tabulated in Ref[13], one can easily calculate the elements H_{rs}^{ea} defined in Ref.[12, 13]

$$H_{rs}^{ea} = \frac{2N_e}{3}\sqrt{\frac{8\varkappa T}{\pi m}} \sum_k (-1)^k \beta_k^{(r,s)} \left[\frac{a\,\Gamma(k+3)}{(1+bT)^{k+3}} + C\,T^d \frac{\Gamma(k+d+3)}{(1+fT)^{k+d+3}}\right], \quad (5)$$

where Γ is the Euler gamma function, and T is the temperature in eV.

Shkarofsky[11] assumed that the kinetic function g_σ, in the case where only, an external d. c. field is present, is a function of $p\ (= \langle \nu_{ea} \rangle / \langle \nu_{ei} \rangle)$ only. However this assumption is valid in cases where Q_m does not change rapidly with energy. Namely, in the case of Ar, Kr and Xe plasmas g_σ depends directly on temperature[19].

For evaluation of electrical conductivity using $\sigma = \frac{N_e e^2}{m \langle \nu \rangle} \frac{1}{g_\sigma}$ [11] one has to know the electron density and its dependence on temperature. The electron density, for given temperature and initial pressure, was calculated from the Saha equation with the Unsöld estimate of the lowering of ionization potential[20] due to the presence of electrons in the plasma

$$\Delta E = 6.96 \cdot 10^{-7} \sqrt[3]{Ne} \quad (\text{in } eV). \quad (6)$$

Nonideal plasma. In calculating the nonideality effects one should take into account the fact that two-particle correlation is not negligible in the interval between two consecutive collisions, that is, the influence of interaction between two particles can not be confined to the act of collision itself. Consequently the form of the Botzmann collision integral is modified[8]

$$I^{\text{nonid}} = \left\{1 + \frac{1}{\pi}\left[\int_0^\infty \cos \vec{k}\vec{w}\,\tau \cdot \tau\, d\tau\right] \frac{\partial}{\partial t}\right\} I^{\text{id}}. \quad (7)$$

Such a collision integral cannot be evaluated directly, because of the divergency in the correlation term. However, some approximations are possible. If we substi-

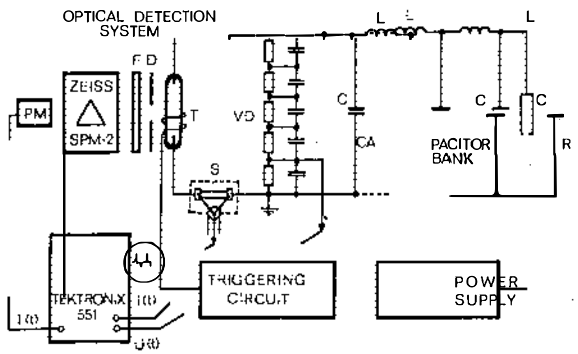


Fig. 2. Experimental arragement.

tute $\tau = r/w$, where r is the interparticle distance and $\vec{k}\vec{w} = w/r_0$ (where $r_0 = e^2/\varkappa T$, and w is the relative velocity), the integral (7) becomes

$$I^{\text{nonid}} = \left\{1 + \frac{1}{\pi w^2}\left[\int_0^\infty \cos\frac{r}{r_0}\cdot r\,\mathrm{d}r\right]\frac{\partial}{\partial t}\right\} I^{\text{id}}. \tag{8}$$

As the correlation can be significant only at $r < \lambda$ (where λ is the mean free path) the following approximation appears to be the most suitable

$$I^{\text{nonid}} = \left\{1 + \frac{1}{\pi w^2}\left[\int_{r_0}^{\lambda} \cos\frac{r}{r_0}\,\mathrm{d}r\right]\frac{\partial}{\partial t}\right\} I^{\text{id}}. \tag{9}$$

Furthermore, if we substitute $\partial I/\partial t \sim I/\tau_0 = wI/\lambda$, the collision integral for a nonideal gas takes the following approximate form

$$I^{\text{nonid}} = \left\{1 + \frac{1}{\pi \lambda w}\int_{r_0}^{\lambda} r\cos\frac{r}{r_0}\,\mathrm{d}r\right\} I^{\text{id}}. \tag{10}$$

This correction was introduced for all three types of collision (e-i, e-e, e-a) which were taken into account in the calculation.

In calculating the collision frequency for e-i encounters the model collision integral proposed by Klimontovich[9] was used. Namely, this author assumed a collision model in which one of the particles is at rest and acts with a screened potential $\varphi_{12}(r) = (e_1 e_2/r) \cdot \exp(r/r_D)$ (in the LTE approximation), whereas the other particle acts with a Coulomb potential $\varphi_{21} = e_1 e_2/r$. This model is similar to the assumption that only electrons contribute to the screening (ions are too slow to follow an electron in its motion), which was introduced by Devoto[6]. The collision integral obtained in such a way converges at large distances, and the»Coulomb logarithm« changes its value[9].

The local equilibrium distribution function, f_0, assumed to be Maxwellian in the ideal plasma calculations, was calculated following the recommendations of Kaklyugin and Norman[10], where the correlation between the charged particles was taken into account as well. A correction for the change in the value of the external electric field, caused by the deviations in the mechanism of interaction, was also taken into account. Klimontovich[21] found that, as a consequence of the lowering of the external electric field strength, the value of electrical conductivity was lower by a factor of $1 - 1/12 n_D$, which is a considerable correction when $n_D \sim 1$.

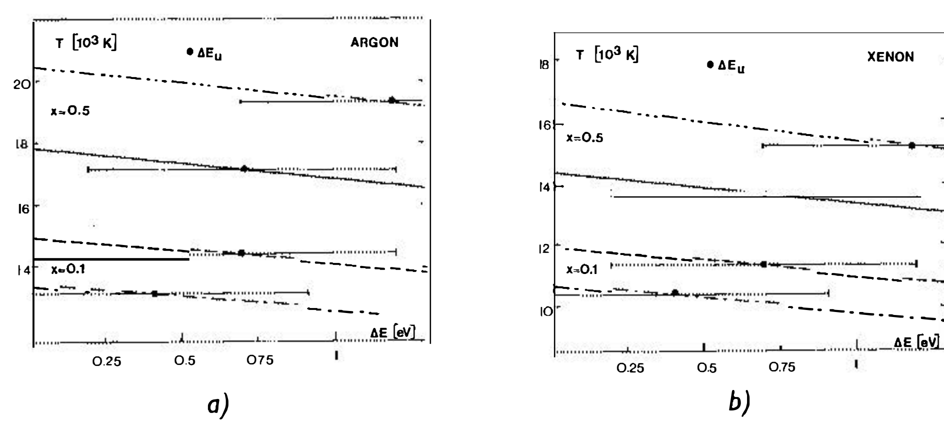


Fig. 3. Variation of temperature calculated from the Saha equation (for given initial pressure and ionization degree) with lowering ionization potential.

— · · · — $p_0 = 500$ torr, $x = 0.5$, ⋯⋯——— $p_0 = 100$ torr, $x = 0.5$

— — — — $p_0 = 500$ torr, $x = 0.1$, — · — · — $p_0 = 100$ torr, $x = 0.1$

a) Argon b) Xenon

Determination of the degree of ionization is also a crucial point of the calculation. Namely, in our concept of calculation the temperature is given as an independent variable, and the corresponding degrees of ionization are to be evaluated using an equation for equilibrium composition. For that purpose, the Saha equation has been used, with a correction introduced in the form of an effective lowering of the ionization potential. Unfortunately, the lowering of the ionization potential has been exactly calculated for a fully ionized, hydrogen plasma of the Debye type only[22]. For a non-Debye plasma it seemed more appropriate to use the Unsöld estimate[20].

Nevertheless, we examined how much an uncertainty in the value of ΔE may influence the calculated values of electrical conductivity. For that purpose, we calculated electrical conductivity for given values of ΔE. The results given in Fig. 1 show that an error of $\pm 0.5\ eV$ in ΔE can cause an error from 2 to 15% in the electrical conductivity.

3. *Experimental*

Our measurements were carried out on a pulsed arc in argon and xenon. There are two main reasons for such a choice. First, the homogeneity of pulsed arcs in heavy rare gases due to the large contribution of radiation to its energy balance considerably simplifies the problems concerning an exact determination of radial temperature distribution. Second, electron densities up to $5 \cdot 10^{18}\ cm^{-3}$ can be obtained in a high current pulsed arc, hence this is, perhaps, the simplest way to create the conditions for occurrence of dense plasma effects.

The pulsed arc was produced in cylindrical tubes 10 cm long and with an inner diameter of 8 mm. The tubes were filled with argon and xenon at initial pressures in the range from 20 torr to 500 torr. The basic element of the discharge circuit was a capacitor bank composed of 6 LC elements. The battery was designed to give a rectangular pulse, when the discharge tube is an element of the circuit[23]. The experimental set-up for the production of the plasma is given in Fig. 2. In the plasma created in the pulsed arc the number of particles in a Debye sphere was $n_D < 2.6$, hence it was of a non-Debye type.

Electrical conductivity was determined from the Ohm's law in the stationary part of the discharge

$$\frac{I}{E} = 2\pi \int_0^{r_0} \sigma(r)\, r\, \mathrm{d}r, \tag{11}$$

where I is the discharge current, E the electric field strength, and r_0 the radius of the tube. From the above equation the value of the axial electrical conductivity, $\sigma(0)$ was determined using a standard iterative procedure (see for instance Ref.[3]) where a relation between the average electrical conductivity $\sigma_m = I/Er_0^2\,\pi$ and the average temperature T_m was taken as the starting function $\sigma(T)$. With the function $\sigma(T)$ adopted in such a way, and the known temperature distribution[24] $T(r)$

$$I(r) = \begin{cases} I(0) & r < \dfrac{r_0}{2} \\ 1.18\, T(0)\left[0.5 - \dfrac{r}{2r_0}\right]^{0.11} & \dfrac{r_0}{2} < r < r_0 \end{cases} \tag{12}$$

the values of $\sigma(0)$ were determined using equation (11). $T(0)$ was determined from the relation

$$T_m = \frac{2}{r_0^2} \int_0^{r_0} T(r)\, r\, \mathrm{d}r. \tag{13}$$

Then the relation between $\sigma(0)$ and $T(0)$ was determined again and the whole procedure repeated. The $\sigma(0)$ obtained after third iteration was taken as the experimental value of electrical conductivity.

The temperature T_m was determined from the Saha equation

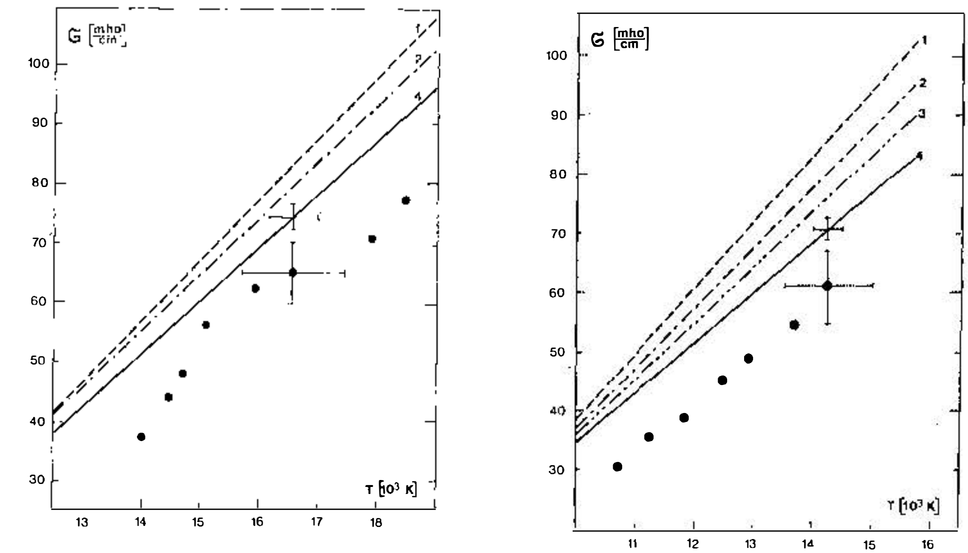
$$\frac{N_{em}^2}{N_0 - N_{em}} = 2\,\frac{U_1(T)}{U_0(T)} \left(\frac{2\pi m_0 \varkappa}{h^2}\right)^{3/2} T_m^{3/2} \exp\left(-\frac{E_{ion} - \Delta E}{\varkappa T_m}\right), \tag{14}$$

where the average electron density was measured by laser interferometry method. Such a simple equation of equilibrium composition is determined by the comparatively low temperature of the discharge and by the fact that the conditions for the establishment of LTE in heavy noble gas plasmas at high densities ($N_e > 10^{17}$ cm^{-3}) are fulfilled and all the components have the same temperature. The equilibrium parameter α[3] has a value $\alpha > 10^4$ in case of xenon high current discharge, and $\alpha > 100$ in case of argon. Hence the error due to the assumption that all the components of the plasma have the same temperature is negligible.

The definition of the lowering of ionization potential ΔE_1 is another crucial point of this method. The value of this perturbation factor was exactly calculated for Coulomb interactions only, and for a fully ionized hydrogen Debye plasma ($n_D \gg 1$).

It seems that the estimate of ΔE_1 on the basis of the nearest neighbour approximation[20] is more acceptable in the case of nonideal plasmas ($n_D \sim 1$). In order to estimate the error due to the uncertainty in the value of the lowering of ionization potential we calculated the temperature making use of (14) for various values of ΔE_1 keeping the degree of ionization constant. The results of this calculation (given in Fig. 3) show that an uncertainty of $\pm$ 0.5 eV in the value of ΔE_1 may produce an error from 5 to 7.5% (Xenon) or 3.5 to 5% (Argon) in the value of temperature. This error combined with that made in the meausurements of electron density by laser interferometry method gives a total error in the determination of temperature T_m of about 10% (Xenon) and 15% (Argon).

The results for discharges in Argon and Xenon at initial pressure $p = 200$ torr are shown in Fig. 4. They are presented as a plot of temperature versus electrical conductivity. The results of calculations with and without nonideality effects, together with estimated errors in calculation and measurement, are also given in Figs. 6 and 7. The calculation errors are due to the uncertainties in the fitting of the experimental data for electron-atom momentum transfer cross-sections and to the uncertainties in the equilibrium composition of the plasma.



a) b)

Fig. 4. Temperature dependence of electrical conductivity. Solid circles represent experimental values. Calculations: 1 — for an ideal plasma, 2 — following recommendations in Ref.[8], 3 — following recommendations in Refs.[8, 9] 4 — following recommendations in Refs.[8, 9, 10].

a) Argon $p_0 = 200$ torr, and b) Xenon $p_0 = 200$ torr.

4. *Conclusion*

The inclusion of nonideality effects in the calculations of the electrical conductivity of heavy noble gas plasmas provides considerable corrections to their values. However, the discrepancies between experimental and theoretical results still exceed the estimated errors. There are many possible sources of these discrepancies.

First, in the present calculations only electron-electron, electron-ion and electron-atom elastic collisions have been taken into account. The contribution of ions to the energy transport as well as the contribution of nonelastic collisions have been neglected, the latter without any preliminary estimates.

Second, classical kinetic equations do not take into account the encounters at small distances, which become more frequent at high densities.

Third, the effect of electron trapping by ion-acoustic waves (which has been studied in collisionless plasmas[25, 26]) may also occur in high density plasmas.

More sophisticated calculations, which would provide more exact values for electron-atom momentum transfer cross-sections, may turn out to be necessary. More accurate electrical and temperature measurements would reduce the experimental errors even more. Furthermore, since at the present state of the art a good experiment may yield more reliable values for electrical conductivity than the available theory, more accurate experimental data should enable one to make a better choice between these theoretical models, or to suggest a new one.

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PROUČAVANJE ELEKTROPROVODNOSTI U PLAZMI TEŠKIH INERTNIH GASOVA

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Sadržaj

Izvršeno je uporedno eksperimentalno i teorijsko proučavanje elektroprovodnosti guste plazme teških inertnih gasova. Izvršeni su proračuni prema postojećim teorijama i pokazano je da postoji sistematsko odstupanje između proračunatih i eksperimentalno dobijenih vrednosti za elektroprovodnost.