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Anomalous Doppler broadening of Hydrogen lines due to excitation by fast neutrals in low pressure Townsend discharges

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Abstract. For many years weak Doppler broadened wings were observed in hydrogen lines emitted from low pressure discharges. Explanations were usually related to dissociative processes or excitation by ions. Petrović and Phelps were the first to perform the measurements in Townsend discharges and by observing the emission along the axis of the discharge two groups of fast particles were observed one going towards the cathode and one away from the cathode. Current dependence ruled out excitation of the fast atoms produced in dissociative charge transfer collisions by electrons. Thus the results could only be explained by excitation by fast neutrals produced either in charge transfer collisions or by neutralization and reflection of ions at the surface. The energies of up to the total available energy (potential fall) were found, though the reflected component had typically 3 times smaller energy. Even more pronounced effects were found at lower E/N in mixtures of hydrogen and argon and methane and argon, though similar effects were observed with other light rare gases. These effects as found in Grimm discharges were studies in great detail by Konjević and coworkers. In addition, some implications for suspected cold fusion were recently analyzed in the literature.
We shall, however, discuss the implications of these processes in plasma etching of integrated circuits. Also some preliminary results of Doppler profiles calculated by using Monte Carlo simulation technique will be shown.

Key words. Low current Townsend discharge, dissociative excitation, fast neutrals, Doppler shift, plasma etching

1. Introduction

The atomic lines emitted from hydrogen discharges are usually the result of the dissociative excitation (Lamb Jr. & Ratherford 1950). Therefore one may expect a broad line profile due to high energy of fragments of dissociation produced by the transition to the repulsive potential curve (Ogawa & Higo 1979, 1980). As a matter of fact Doppler broadened lines from the electron beam excitation experiments of low pressure gas targets have been used to determine the energy distribution of fragments and consequently to correct the cross section measurements (Tasić et al. 1989). Similar Doppler broadened profiles have been

Send offprint requests to: Z. Petrović observed for other dissociative processes such as dissociative recombination and ionization.

At the same time anomalously broadened profiles have been found in a number of gas discharges including DC and RF plasmas. Under the term anomalous we mean that the broadening goes beyond that defined by the gas temperature and in due course we shall see that it often goes beyond the broadening determined by energies of fragments from dissociative processes defined by molecular potentials. We will show the development of the ideas that led to the explanation of the anomalous broadening and we will try to give further and more recent developments of the applications where Doppler broadened line profiles may be used to provide diagnostics and understanding of the application.

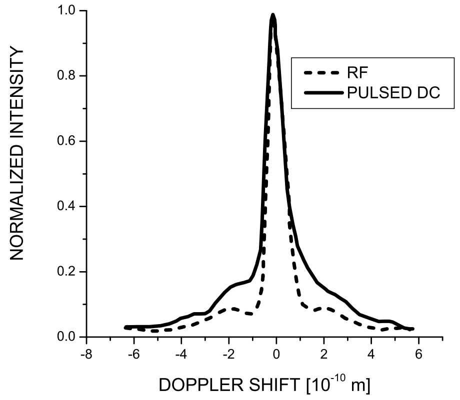

Fig. 1. Doppler broadened profiles of Hβ line in pulsed DC and RF discharges in hydrogen (Vrhovac et al. 1991a,b).

1.1. Early Observations in DC and RF discharges

Anomalously broadened hydrogen profiles were observed in a number of glow discharges in hydrogen. First systematic study was performed by Benesch and coworkers (Benesch & Li 1984; Li Ayers and Benesch 1988). They have shown a difference between the observation along the direction of the electric field and perpendicular to it. Similar results were found by other groups including the group at the Institute of Physics in Belgrade (Bzenić et al. 1990). Most puzzling result was the presence of the wings that indicated motion away from the cathode that could not be explained by fast ion acceleration towards the cathode. Side on observations revealed symmetric but broadened line profiles. In both cases the energies of fragments of more than 10 eV were detected.

Similar results were obtained in RF discharges (Capelli et al. 1985; Baravian et al. 1987; Vrhovac et al. 1991a,b). Most importantly these results were associated with plasmas that were to be used in processing of integrated circuits. An example covering both pulsed DC and RF discharges is shown in Fig.1. In DC the wings are higher and obviously more asymmetric.

Several explanations were offered. One group was based on dissociative processes such as dissociative excitation, ionization and even recombination (Capelli et al. 1985). The process in question is certainly present and contributes to the line profile yet most discharges provided wings with energies in excess of those defined by the molecular potential curves. The fast fragments with much higher energies were in particular observed in the end on measurements in DC discharges (Benesch & Li 1984; Bzenić et al. 1990). These observations led to the discussion of possible dissociative excitation by fast ions that would occur in the sheath of the glow discharge. While this model gained support it could not explain an almost equally abundant wing due to particles moving away from the cathode.

However, it should be noted that Zdenko Šternberg (Šternberg 1988) has made an analysis of the anomalous broadening much before most of the groups mentioned above. His inspiration came from the fusion research but his work was aimed at glow discharges. The idea was that the ions performed dissociative excitation resulting in the wing with velocities towards the cathode due to efficient momentum transfer from heavy particles. It was proposed that the excited particles moving in the opposite direction were generated first as fast neutrals by neutralization and reflection from the surface of the incident ions. In the next step the fast neutrals were excited by electrons in the glow region. If this proposal is correct the first wing would be proportional to the current while the second wing would be proportional to the square of the current as it involved two electron induced steps, ionization and subsequent excitation.

To conclude we may state that apart from dissociative processes that would give fragments of up to approximately 15 eV and peaking at around 5 eV the profiles observed in glow discharges contained a component that had much higher energies. While one could perhaps justify such energies of fragments by the momentum transfer between ions and target molecules one had difficulty explaining the wing due to particles moving away from the cathode. It is the combination of these effects that may be labelled *anomalous*.

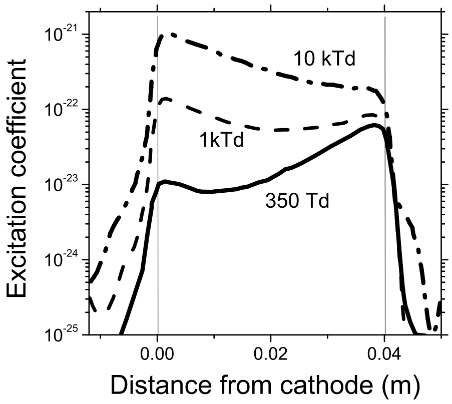

Fig. 2. Spatial emission profiles absolutely calibrated to the excitation coefficients for Townsend discharges in hydrogen(Petrović & Phelps 1991a).

1.2. Kinetics of Fast Heavy Particles in Low Current Discharges

In mid 80s Phelps (with coworkers) has moved the boundary of low current (swarm) experiments in Townsend discharges to lower pressures and higher values of E/N in pursuit of non-hydrodynamic effects. However, they discovered that excitation under those circumstances appeared to be generated by fast heavy particles and instead od the expected exponential growth of emission towards the anode (which is typical for electron excitation) they observed growth of emission towards the cathode (Jelenković & Phelps 1987; Phelps 1994). This was explained by the growth of the flux of fast neutrals which were shown to be much more efficient in excitation than the fast ions. The fast neutrals are produced by charge transfer collisions (Phelps et al. 1987).

This work gave an indication that it is actually the fast neutrals that perform dissociative excitation and give momentum to the fragments. These results also gave the basis for quantitative comparisons based on models that could be verified against swarm experiments that have a well defined field profile and high accuracy of the well defined observables.

As a result of both of these lines of work an idea was developed to study the line profiles from the Townsend discharges in hydrogen at very high E/N. The advantage of such a setup is that the field in low current discharges is quite uniform and not affected by the value of the current. Thus an exact model may be developed which would allow quantitative comparisons.

2. Swarm experiment of Petrović and Phelps

Measurements of spatial emission profiles in pure hydrogen were performed in a standard drift tube (Jelenković & Phelps 1987) and those were confirmed by the results from Belgrade (Stokić et al. 1992). Three examples of the spatial profiles observed in pure hydrogen for different E/N are shown in Fig.2 (Petrović & Phelps 1991a).

One electrode in the system was made of vacuum grade graphite and the other was a thin film of gold-palladium (Au-Pd) which was transparent and allowed end on measurement along the direction of the electric field. Even though very small currents of the order of 1 μA over the electrodes of the diameter of 10 cm were used, it was possible to record line profiles (of Hα). Resolution could not be very high but it was sufficient to reveal very asym-

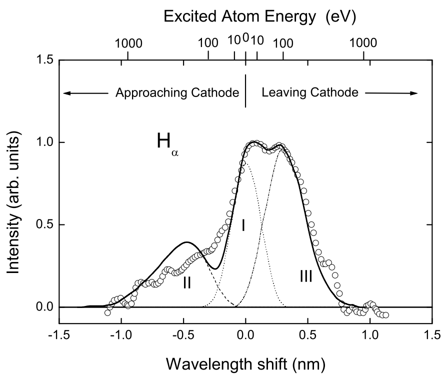

Fig. 3. Doppler broadened Hα profile at 10 kTd from a discharge in hydrogen with AuPd cathode (Petrović et al. 1992).

metric Doppler broadened profiles (Petrović & Phelps 1989, 1990; Petrović et al. 1992)

In Fig.3 we show a profile obtained at 10 kTd in hydrogen. Applied voltage was around 2000 V and the gas pressure was 20 Pa. The profile consists of three parts, the central line is due to dissociative excitation by electrons and it is determined by the instrumental resolution though its width is consistent with the typical energies of dissociated fragments which are of the order of 5eV-10 eV. The wing with fragments approaching the cathode is broader of the two wings but somewhat smaller. It extends all the way to 1000 eV which indicates that there are ions that make no more than one collision in the discharge and thus may attain very high energies of the order of a half of the maximum available energy. The quantitative comparison with a model (dashed lines) indicates that the direct contribution of ions to the dissociative excitation is very small but the fast neutrals produced in charge transfer collisions are able to provide the required level of excitation (and Doppler broadening).

Numerous attempts were made (Petrović et al. 1992) to check different possible excitation scenarios of the group of fragments going away from the cathode. Finally, it turned out that the best quantitative agreement was given with the fast neutral particles reflected from the cathode and doing subsequent excitation. This flux will include both reflected fast neutrals and the fast neutrals formed at the surface by neutralization of beams of ions incident on the surface. Therefore it is possible that the integral of the wing due to reflection exceeds that of the direct excitation.

This explanation is different from Šternberg's idea described above as it does not require excitation by electrons but the fast neutrals actually perform dissociative excitation. At the time when Šternberg developed his model the role of fast neutrals was not understood in the literature. In addition both wings were proportional to the current density and not to its square. Experimental tests and calculations of profiles were made to check the surface excitation hypothesis and the lifetime was too short to give a satisfactory agreement with the experiment. In Fig. 4 we show spatial profiles measured and calculated indicating that the hypothesis that returning fragments are excited at the surface in the process of neutralization is not valid. The difference between the absolute intensity of emission between scans for graphite and AuPd cathodes shows the difference due to reflected fast neutrals since reflection from the graphite is very small.

Absence of reflected component for graphite cathode may be best seen from Fig.5 where we show line profiles with both electrodes (Petrović & Phelps 1991a; Petrović et al. 1992).

The proposed explanation of anomalously broadened profiles obtained from the low current (swarm conditions) discharges measurements could be tested directly on the basis of a simple beam like model (Phelps et al. 1987). It also gave first quantitative tests of the proposed models and thus it provided the foundation for explaining similar results in more complex higher current discharges.

2.1. Measurements in other gases and gas mixtures

Similar results for the spatial emission profiles of Hα were found in methane and it is expected that it would be found in (Petrović & Phelps

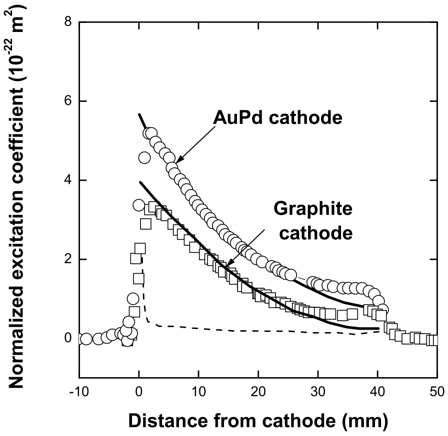

Fig. 4. Spatial profiles of Hα emission at 10 kTd from a discharge in hydrogen with AuPd and graphite cathodes. For comparison we show a dashed line indicating the expected profile for surface excitation(Petrović et al. 1992).

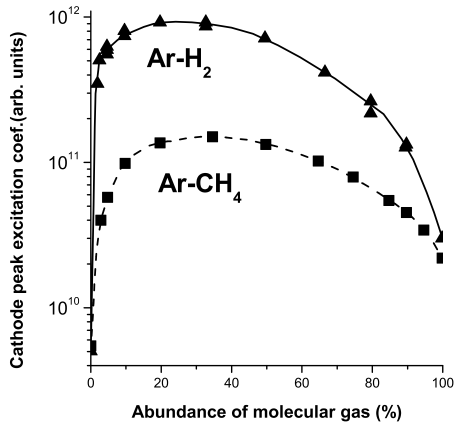

Fig. 6. Excitation coefficients at the cathode as a function of the abundance of molecular gas in argon (for hydrogen and methane mixtures with argon) (Jelenković et al. 1990; Petrović & Phelps 1991b). Measurements were made at 1 kTd at 150 mTorr and V=190 V for hydrogen and 2 kTd, 100 mTorr and 260 V for methane mixtures.

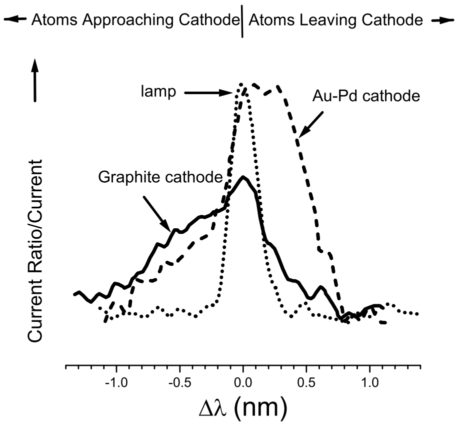

Fig. 5. Line profiles of Hα emission at 10 kTd from a discharge in hydrogen with AuPd and graphite cathodes (Petrović & Phelps 1991a; Petrović et al. 1992).

1991a; Šašić et al. 2004) all other hydrocarbons.

In order to see the anomalous Doppler broadening, in pure hydrogen, as mentioned above, one needs to go to very high E/N beyond several kTd with the available energies in excess of 1000 V. However, it was found that the effect of fast neutral excitation has been observed at relatively low available energies of the order of 200 V for mixtures of argon and hydrogen or methane. The peak close to the cathode that indicates heavy particle excitation increases dramatically for small abundances of either hydrogen in argon or argon in hydrogen and the same thing was found for methane argon mixtures as can be seen in Fig. 6. A similar explanation to that in pure hydrogen may be developed with a possibility that excitation may be achieved either by fast argon or fast hydrogen atoms. Doppler broadening in argon hydrogen mixtures was more difficult to detect due to smaller energies but some degree of broadening consistent with the energies involved was observed. In any case, not surprisingly, most of the observations of the effect of fast neutrals on excitation of Balmer lines were detected in argon hydrogen mixtures (Petrović & Phelps 1991a).

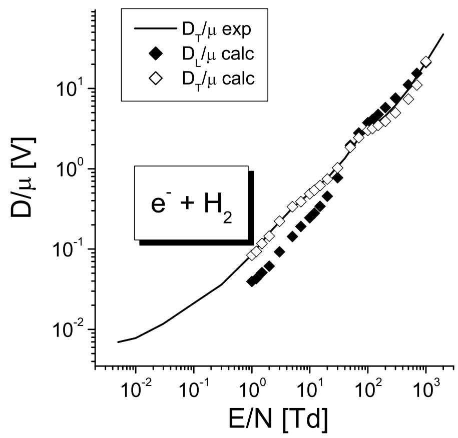

Fig. 7. Comparison of calculated and experimental data for D_T/μ for electrons in hydrogen indicating that the set of cross sections is yielding good electron energy distribution function. Calculated results for D_L/μ are also shown.

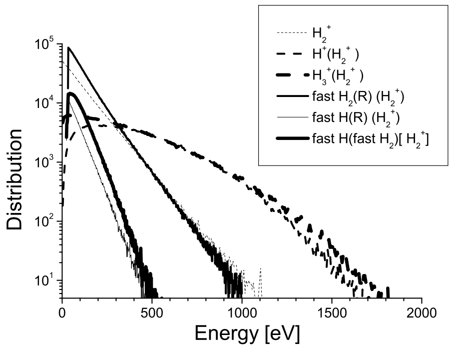

Fig. 8. Energy distribution of fast ions and fast neutrals in a hydrogen discharge at 10 kTd. The results are consistent with Doppler profiles and fast neutral excitation.

3. Monte Carlo simulation of Doppler profiles in Townsend discharges

The numerical model used by Phelps and coworkers (Phelps et al. 1987) is a simplified beam like model. Exact calculations for such conditions have never been performed for hydrogen. In this paper we will show the first preliminary results for Monte Carlo calculations of the transport of all particles in pure hydrogen. A code based on null collision technique has been developed to include electron, ion and fast neutral particles in the background gas (Petrović & Stojanović 1998; Stojanović & Petrović 1998; Stojanović et al. 1997). The code has been tested on nitrogen and argon and the results were found to be in a very good agreement with the experimental data.

A set of well tested cross section data has been applied for electrons, ions and fast neutrals (Phelps 1990, 1992). One example of a good agreement of predictions for electrons is shown in Fig.7. A similar and even better agreement was achieved for other transport coefficients. In particular it was important to have good predictions for the ionization coefficient. As for fast ions and neutrals calculations were performed coupled to electron simulations. The energy distribution functions for fast neutrals and ions were sampled as shown in Fig.8.

Fast neutrals and ions collide with gas phase molecules and induce dissociative excitation. The components of excited fragments were sampled under simplifying assumptions and a simulated line profile was obtained. The results for the wings are shown in the Fig.9 and they agree reasonably well with the experimental results shown in Fig.3. These results also include contributions of different processes, but the reflection at the cathode was taken to be 100%. Adjustment of reflection is possible to fit the experimental data but also one may adjust other parameters such as energy losses at the surface and the efficiency of neutralization.

It is very difficult to state that the Monte Carlo model has confirmed the calculations based on the beam model of Phelps and coworkers but certainly it provides more flexibility that allows both modelling of experiments with strong fast neutral component and also it is possible to use them to obtain quantitative data for different processes.

4. Recent observations and applications

Study of Doppler broadened profiles in glow discharges were conducted by Barbeau & Jolly

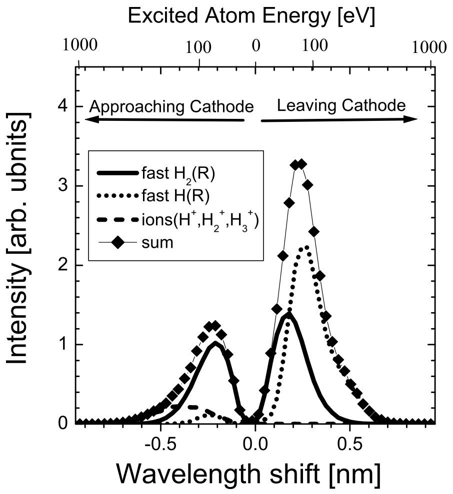

Fig. 9. Balmer Hα line profile obtained from the Monte carlo simulation in a hydrogen discharge at 10 kTd (all conditions were the same as in the actual experiment).

(1990) at the same time as Petrović and Phelps performed their measurements. They have considered a possibility that the wing of the profile due to particles moving away from the cathode is due to surface excitation of ions that are neutralized at the surface and reflected as excited species.

The most systematic study of line profiles affected by fast neutral dissociative excitation came from Konjević and coworkers. They have studied the so called Grimm discharge where they followed effects both in pure hydrogen and its mixtures with rare gases(Konjević & Kuraica 1992; Videnović et al. 1996; Gemišić Adamov et al. 2004) and including the studies of the effect of the cathode material on Doppler broadened line profiles (Gemišić Adamov et al. 2004). While the field distribution in their system is quite complex and there are no accurate models the basic phenomenology and explanation of the results observed in those experiments is the same as that proposed by Petrović and Phelps with the addition of the complexity introduced by sputtered material and nonuniform fields. These authors have extended their studies to measure Doppler profiles from microwave discharges (Jovićević et al. 2004) in order to disprove recent claims that the efficient fast neutral excitation in rare gas hydrogen mixtures may be taken as a proof of cold fusion (Mills et al. 2002).

The effect of fast neutrals has been considered for a number od DC glow discharges. Most systematic study was performed for argon by observing spatial profiles and comparing them to the calculations by a hybrid model (Marić et al. 2003; Donko 2000). A study of spatial profiles in argon silane mixtures (Matsuda et al. 1994) showed a large effect perhaps in a similar way that the effect was found to be enhanced in argon methane mixtures by Petrović and Phelps.

Doppler broadening may be observed in Optogalvanic signal as a function of the wavelength in studies of cathode fall by (Doughty et al. 1985). These studies were associated with the measurements and modelling (Revel et al. 2000) of gas heating by a gas discharge.

Monte Carlo simulations of heavy particle effects for discharges in nitrogen were performed for a given (not calculated in a self consistent manner) electric field profile (Yu et al. 2001). Simulations in pure hydrogen were also performed but those were mainly focused on ion transport (Bretagne et al. 1994), but hybrid codes including heavy particle Monte Carlo simulations were performed for pure hydrogen and for hydrogen argon mixtures (Bogaerts & Gijbels 2000a, 2002b,c).

Doppler broadening is often used in diagnostics of fusion plasmas (Helium line broadening due to the presence of fast neutrals is shown in Fig.10 (Bogen et al. 1989). This work may also be of relevance for very high E/N discharges that are used for practical applications such as pseudo spark switches (Glišić et al. 1998) .

Studies of anomalously Doppler broadened line profiles in RF plasmas include the work of Radovanov et al. (1995) who have studied the time resolved Doppler broadened lines. Anomalously broadened lines from RF discharges have been detected and analyzed by Radovanov et al. (1995); Djurović & Roberts (1993) Balmer radiation has been used in RF discharges to establish the presence of high energy hydrogen neutrals (Gans et al. 2002). In

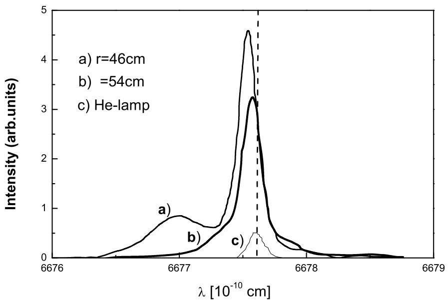

Fig. 10. He singlet line (667.8 nm) profiles from a fusion reactor discharge in helium (Bogen et al. 1989) at two positions (a-b) of the limiter and also including a spectral He lamp (c).

addition, it has been used to study the changes of modes of an RF discharge with dust particles (Stefanović et al. 2003).

The studies and modelling of dc Townsend discharges performed at low pressures may be of relevance for studies of the ion to fast neutral beam conversion (Shimokawa & Kuwano 1992).

5. Fast neutrals in plasma etching

The role of fast neutrals in plasma etching has been studied by Sommerer & Kushner (1991) as a possible source of isotropic etching. It was found that the flux of those fast neutrals is similar to the flux of the ions.

On the other hand applications of specially developed neutral beams were considered. Different applications of fast neutral beams were developed in plasma processing. For example in order to reduce UV photons and contamination from sputtering and to improve the uniformity of the beam multiple reflection system was used to produce hyperthermal neutral beams from ion beams (Goeckner et al. 1997). In general, sources of fast neutrals that are based on mechanical devices (such as choppers) or employ gas expansion through the nozzle suffer from limited energy that may not be appropriate for etching of dielectrics. On the other hand charge exchange of ions from a plasma yields high energy beams efficiently. Surface neutralization of ion beams was also found to be very efficient and able to cover wide range of energies. Efficient fast neutral etching has been demonstrated by using a 4.8 eV neutral beam (Giapis et al. 1995).

In view of the recently developed explanation of the role of the fast neutrals it was proposed that fast neutral etching with energies comparable to those of ions may be feasible (Petrović & Phelps 1991b). The proposal was repeated several years later (Petrović & Stojanović 1998) having in mind treatment of dielectrics and removal of possible causes of defects due to charging. In particular the grid replacing the cathode was suggested as a possible source of fast neutrals for etching (Scott & Phelps 1991).

Independently of our proposal a similar proposition was (Samukawa et al. 2002; Panda et al. 2001) made but etching by fast neutrals has been demonstrated and shown to avoid defects due to charging (Matsui et al. 2001).

6. Conclusions

In this paper we have reviewed some of the evidence for heavy particle excitation and ionization in gas discharges. In particular anomalous Doppler broadening of Balmer lines of hydrogen have been explained. It was found that fast neutral excitation can explain both wings of the Doppler broadened lines. These processes become significant at high values of E/N, i.e., when low values of gas pressure are combined with high applied voltages. Another point where the models including heavy particle excitation can be used is to explain the far wings of the Doppler profiles of Hα radiation in different discharges. Thus the Doppler broadened wings may be taken as an indication of the presence of fast neutrals in discharges containing hydrogen. In discharges normally used for processing of integrated circuits, fast neutrals may be used for plasma etching to avoid charging damage.

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