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N80-33438

TDA Progress Report 42-59 July and August 1980

A Study of the Processes in the RF Hydrogen Gas Dissociator

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The role of the RF gas dissociator in the hydrogen maser is examined. Based on collisional and plasma transport processes, the performance of the source is investigated. It is found that while the complexity of the collisional processes in the RF dissociator prohibits an easily obtained quantitative expression for the performance of the source, it is nevertheless possible to make general inferences concerning the qualitative performance based on collisional effects. An analytical expression for the efficiency of the source in atom production is obtained based on plasma transport processes. On the basis of this study some recommendations are made for the development of more efficient RF hydrogen gas dissociators for use in masers.

I. Introduction

The performance of the hydrogen gas dissociator has a significant effect on the operation of the hydrogen maser. The dissociator affects the performance of the maser in terms of power output and stability by influencing the flux of the hydrogen atoms (Ref. 1) which radiate through stimulated emission; it also affects the operation of the vacuum pumps, thereby indirectly influencing the operational life of the maser itself.

The most significant effect of the dissociator on the H-maser operation, however, is its direct influence on maser breakdown; experience has shown that the degradation of the dissociator performance is one of the primary causes of maser failure.

It therefore appears that an evaluation of the performance of the hydrogen gas dissociator would prove quite useful in extension of efforts towards optimization of H-maser operation. In particular, understanding of the processes that take place within the hydrogen source would be a valuable aid in the work toward the development of a more rugged dissociator with an improved performance.

In Section II of this report background information on the RF dissociator is given. Processes within the dissociator are outlined in Sections III and IV. Section V briefly outlines interactions with wall material and Section VI gives the summary.

II. Background

Of the number of atomic hydrogen sources used in various applications (Ref. 2) none appears more suitable for the hydrogen maser than the RF dissociator (Fig. 1). The reason for this is the strict requirements of the maser application on the available sources. Due to the sensitivity of the maser stability to fluctuation in temperature and magnetic field in the environment (Ref. 2), sources which employ high temperatures to produce atomic hydrogen are not well suited. Furthermore, most sources employ either a heated filament or a light source to dissociate the hydrogen molecules and are therefore susceptible to frequent shutdowns necessary to replace or refurbish those elements. This leaves the RF dissociator as the most suitable hydrogen atom source for maser application. Added to the desirable features of long operational life and minimum environmental disturbances of the RF sources are such other attractive characteristics as low power requirement and small size. These latter features are particularly important in the development of spaceborne masers.

The dissociation of molecular hydrogen in all varieties of sources is brought about by inelastic collision of the molecules with other particles. In the case of the RF dissociator, collisions of the molecules with electrons are primarily responsible for atom production.

When hydrogen gas is introduced in the dissociator bulb and the RF power is turned on, any free electrons present in the bulb will oscillate in the RF field. During the course of these oscillations, electrons collide with the molecules, resulting in the production of a number of particles including atoms, ions and other electrons. The new electrons in turn participate in collisions, thereby producing, and sustaining, a plasma discharge.

Although the above outline gives the principle of the operation of the RF source, it neglects many important details of collisions and particle production. It is the study of these other, more detailed processes which would prove of value in the evaluation of the RF dissociator.

III. Collisional Processes in the Dissociator

The rate of any particular reaction between n_e electrons per cubic centimeter and target particles (molecules, atoms, or ions) of density n_i susceptible to the particular reaction in the atom source is determined by the product $(n_i \sigma)(n_e V)$, where σ is the cross section for the particular reaction and V is the relative velocity. Since the mass of the electron is many times smaller than the mass of the target particle, the relative velocity is essentially equal to the electron velocity, if the collision of electrons with other electrons is unimportant in the plasma. When a plasma fulfills certain conditions (Ref. 3), one may consider a certain distribution for the electron velocity. In particular, for the case of the RF dissociator, we consider an average distribution for the product σV, which is designated as $\langle \sigma V \rangle$ and referred to as the rate coefficient for a given reaction. This concept is brought about since the reaction cross section is a function of the reaction energy, or equivalently, of the relative velocity of the particles involved. In the case of the RF dissociator, the electrons typically have a Maxwellian velocity distribution, and reaction cross sections are of the order of 10^{-18} cm^2 for those interactions that have the highest contribution to the source processes. It is now evident that the rate of a given reaction is determined by the electron and particle densities as well as the rate coefficient for the particular reaction.

The inelastic collisions of electrons with molecular hydrogen can result in a number of processes with varied products. While simple dissociation, i.e., $e + H_2 \rightarrow H + H + e$, does occur as the result of collisions, other processes such as electron impact ionization ($e + H_2 \rightarrow H_2^+ + 2e$) and excitation, followed by photon emissions ($e + H_2 \rightarrow e + H_2^* \rightarrow e + H_2 + h\nu$) are also quite likely to occur. In general, the collision of molecules with electrons can result in four types of processes (Ref. 4), viz, dissociation, ionization, excitation, and recombination. Although all these processes do occur in the source to various degrees, a number of them occur so infrequently as to have minimal influence on the performance of the source. We can now try to characterize the significance of each of these processes in the source by studying each process individually.

(1) *Ionization.* Although ionization of particles as a result of electron impact is not the desired product in the atom source, it is nevertheless the primary process contributing to the density of the electrons and thereby affecting the reaction rates. Electron impact can ionize the hydrogen molecules in the following ways:

$$e + H_2 \rightarrow H_2^+ + 2e \qquad (1)$$

$$e + H_2 \rightarrow H^+ + H + 2e \qquad (2)$$

$$e + H \rightarrow H^+ + 2e \qquad (3)$$

All these reactions result in the production of electrons which can now participate in interactions of their own. Process (2) is also an important source of atom production, with a relatively large cross section (Ref. 5) (about 2×10^{-18} cm^2). The contribution of each of these processes to the plasma in the source can best be illustrated by considering the processes which can ensue after the ion is produced in process (1). The H_2^+ ion can undergo additional collisions resulting in the following reactions:

$e + H_2^+ \rightarrow H^+ + H^+ + e$ dissociation ionization

$e + H_2^+ \rightarrow H^+ + H + e$ dissociation excitation

$e + H_2^+ \rightarrow H + H$ dissociation recombination

$e + H_2^+ \rightarrow H^+ + H^-$ pair production

Obviously such reactions are allowed only if energetically possible. But even with electron energies compatible with the above processes, the only reactions with a significant cross section are the dissociative excitations which produce an atom in the reaction; all others have relatively small cross sections and therefore can be neglected in this analysis.

Therefore, it is apparent that all ionization reactions (1) through (3) can occur with almost equal probability, creating secondary electrons needed to sustain the discharge. Furthermore, processes (1) and (2) can lead to reactions which themselves are a source of secondary electrons. While it is comforting that secondary electrons are created in these reactions which can enhance the electronic density in the plasma, it is not necessarily desirable to produce the ions. Ions drift from the plasma and onto the walls, causing wall damage which results in the efficiency degradation of the source. This point will be elaborated upon in Section V.

(2) *Dissociation and Excitation*. These two processes are discussed together, since oftentimes excitation of one of the product particles occurs following electron impact dissociation. In fact, while the cross section for dissociation $H_2 + e \rightarrow H + H + e$ is about 6×10^{-18} cm^2 at 15 eV impact energy, the reaction which would leave one of the atoms in an excited state (with a subsequent decay via the H_α emission) has a cross section only an order of magnitude smaller. Electron impact ionization of the hydrogen molecule may also leave one of the resulting H_2^+ ions or the H atom (as the case might be) in an excited state.

The excited atoms and molecules deexcite by emitting photons producing the light associated with the RF plasmas. Traditionally, the color of this emission is used in the evaluation of the performance of the source. A red/violet hue (primarily due to the Balmer H_α production) is interpreted to signal production of atoms, while radiation associated with molecular emission is taken to mean that the power is dissipated in molecular excitation.

We have already seen that atoms may be produced without H_α emission with cross sections comparable to unexcited atom production. In the study of recombination we shall see that molecular emission may result as molecular ions recombine with electrons; i.e.

$$H_2^+ + e \rightarrow H_2 + h\nu .$$

It should now be evident that the evaluation of the source performance by the color of the source, or other technique based on the comparison of the intensity of atomic emission with molecular emission, is only crudely related to the efficiency of the source in atom production.

(3) *Recombination*. This process leads to the production of neutral species as a result of the reaction of ions with electrons. Although electron recombination with an ion is generally regarded as an undesirable process in ion sources, under certain circumstances it may be regarded as a desired process in atom sources. The recombination of ions (density n_i) with electrons (density n_e) in the plasma is characterized by the recombination rate α where $dn_i/dt = -\alpha n_i n_e$.

Various modes of recombination have various rates associated with them, and the rates usually are a strong function of the electron energy. One of the important modes of recombination in the plasma is radiative recombination, where an ion combining with an electron yields a neutral species and a photon, viz,

$$X^+ + e \rightarrow X^0 + h\nu .$$

For the case of the plasma in the RF dissociator, the rate of radiative recombination α_r is of the order of 10^{-13} cm^{-3} s^{-1}. Another mode of recombination is the dielectronic recombination, which describes the process involving the interaction of an ion and an electron yielding a particle in a highly excited state. The excited particle may then be either ionized again or become stabilized by photon emission or by collision with other particles. In the range of the electron energies present in the RF dissociator the dielectronic recombination rate α_d is usually much larger than the radiative recombination rate for moderate electron energies.

Finally, electronic collisional radiative recombination should be mentioned, which may be described by the reaction

$$X^+ + 2e \rightarrow X + e$$

In the case of a plasma fulfilling certain conditions, Bates et al. give the following relation for the recombination of protons (Ref. 6)

$$\alpha_{eer} \simeq 10^{-8} (T_e)^{9/2} (n_e)$$

where T_e is the electron temperature in kelvin.

The dominant recombination process for the case of hydrogen dissociator is the dissociative recombination, where electron impact on a molecular ion results in the dissociation of the molecule. The process is very efficient, especially at low energies where the cross section for the process can be as large as 10^{-13} cm^2. Dissociative recombination, therefore, can yield hydrogen atoms at an appreciable rate, only if the densities of ions and electrons are not prohibitively low. Ion densities are limited not only by the ionization rate coefficients, but by other phenomena such as drift out of the plasma and wall neutralization. In fact, ion drift is usually a much larger source of ion depletion in the plasma than recombination effects.

Other modes of recombination such as neutral collisional radiative recombination, as well as processes involving charge transfer, have comparatively smaller rates and are of less significance in the study of the hydrogen gas dissociator. The study of production of negative ions and larger molecules and their ions, such as H_3^+, which can also be of significance, will be excluded here.

The significance of recombination in the dissociator is now evident. Recombination can be a source of atom production in the dissociator, as well as a source of photons. While the former effect is desirable, the latter effect contributes to the light produced by the hydrogen source, making the consideration of the color an unreliable technique for the evaluation of the performance of the source, as mentioned before.

So far we have considered the collisional processes that occur in the hydrogen gas dissociator. We can now proceed to formulate a procedure for the evaluation of the rate of atom production in the source. Such an evaluation is in principle simple. Essentially, we can evaluate the rate of atom production by summing all the processes that yield atoms and subtracting all processes that result in the loss of atoms. Specifically, dissociation of hydrogen molecules, dissociative recombination of molecular ions and neutralization of protons yield atoms, while ionization and escape of atoms from the source are the primary atom loss mechanisms. For those mechanisms which have known reaction rates such as dissociation of the molecules, or the dissociative recombination of molecular ions, one may calculate rates, assuming the density of all particles are known. Each rate would be equal to $n_t n_e \langle \sigma_{t,e} v_e \rangle$ where n_e is the density of electrons, n_t the density of the target, $\sigma_{t,e}$ the cross section for the particular process, and $\langle \sigma_{t,e} v_e \rangle$ the rate coefficient. The rate of the escape of the atoms can also be computed from the knowledge of the temperature of the gas and the confinement time. Processes such as recombination of atoms at the walls, and other wall effects are a great deal more difficult to quantify exactly.

IV. Plasma Processes

The only laboratory parameters which affect the performance of the dissociator are the pressure of the hydrogen gas in the dissociator, the RF power input, and the dissociation geometry. Ideally, then, we should look for a relationship between the efficiency of the source and these parameters. While in practice such a relationship is quite difficult to obtain, it is nevertheless possible to examine the efficiency of the source in the context of transport phenomena in the source plasma, for approximate cases. Such a relationship will fall short of an exact description. In what follows, we approximate (Ref. 7) the plasma behavior in the RF source with that of a positive column of a dc plasma.

We can start such a description by considering a source which has a cylindrical geometry with respect to the applied electric field $\vec{E}$, assumed to be parallel to the axis. We also make the following assumptions concerning the plasma in the source. We assume that at every point the net charge density is zero, that is $n_e = n_i \equiv n$, and the temperature of the molecules is constant in space and time; we further assume that the plasma properties are homogeneous in the direction of the cylindrical axis and that they are time independent; furthermore, positively charged particles are produced in collisions of ground state molecules with electrons, and no negative ions are formed. Finally, we assume that the diffusion of charged particles onto the walls and their subsequent recombination is the only mechanism responsible for their loss. All these assumptions are realized to various degrees in the RF dissociator and none of them present a severe deviation from the actual case of the source.

We now can write down the following equations (Refs. 8, 9) to describe the distribution of charged particles in the source. Starting with

$$n_e = n_i \equiv n \tag{1}$$

the flux of charged particles with a drift velocity V_d can be written as

$$\phi = nV_d = D_a \nabla n \tag{2}$$

where D_a is the coefficient of ambipolar diffusion. Taking the divergence of Eq. (2) we get

$$\nabla \cdot \phi = D_a \nabla \cdot \nabla n \tag{2}$$

and since

$$\nabla \cdot \phi = \nu_i n$$

where ν_i is the collisional ionization frequency, we have

$$D_a \nabla^2 n = -\nu_i n$$

or

$$\nabla^2 n + \frac{\nu_i}{D_a} n = 0 \tag{3}$$

For a cylindrical geometry we expect that the density of charged particles is only a function of r, the radial distance from the symmetry axis. We therefore have

$$\frac{d^2 n(r)}{dn^2} + \frac{1}{r}\frac{dn(r)}{dr} + \frac{\nu_i}{D_a} n(r) = 0 \tag{4}$$

This equation has a solution of the form:

$$n(r) = n_0 J_0 \left(r \sqrt{\frac{\nu_i}{D_a}} \right) \tag{5}$$

where n_0 is the charge density on the axis and J_0 is the zeroth-order Bessel function. Imposing the boundary condition that the density of charged particles is zero at the wall, i.e., $n = 0$ at $r = R$, where R is the radius of the source, we obtain

$$\sqrt{\frac{\nu_i}{D_a}} R = 2.4 \tag{6}$$

where the numerical function 2.4 is the first root of J_0. Equation (5) gives us the condition for sustainment of the plasma in the source. We can use this equation to obtain T_e, the temperature of electrons in the plasma. To do so, we use the assumption of Maxwellian velocity distribution for electrons so that we may write down the following equation for ν_i (Ref. 9):

$$\nu_i = A\, p\, V_i^{3/2} \left(\frac{eV_i}{kT_e} \right)^{-1/2} \exp(-eV_i/kT_e) \tag{7}$$

where A is a constant characteristic of the particular gas, V_i is the ionization potential of the gas, and p is the pressure. We can write the approximate relation:

$$D_a = \frac{kT_e}{e} \mu^+ \tag{8}$$

which is valid when the electron temperature is much larger than the ion temperature (assumed to be approximately the same as the ambient temperature), with μ^+ as the mobility of the ions. Combining Eqs. (6), (7) and (8), we get

$$\frac{\exp(eV_i/kT_e)}{(eV_i/kT_e)^{1/2}} = 1.2 \times 10^7 (CpR)^2 \tag{9}$$

where C is a characteristic constant of the gas and is approximately equal to 10^{-2} in the case of H_2 (Ref. 10). Equation (9) permits us to calculate the temperature of the electrons in the plasma, when the pressure p and the radius of the source R are known. For example, for a typical source with $R = 2.5$ cm and $p = 0.1$ torr, we have in the case of hydrogen gas an electron temperature of about 35000 K. The important point of Eq. (9) is that there is essentially a relationship between T_e/V_i and the product CpR. In other words, for a given gas we have a relationship between T_e and the product pR, which can be used to obtain a desired T_e by varying p and R.

We next consider the potential at the vicinity of the wall arising from the diffusion of the charged particles. For an insulating wall a (Debye) sheath is produced at the wall to insure that the number of (faster moving) electrons arriving at the wall is equal to the number of (slower moving) ions at the wall. It can be shown that the potential across this sheath is given by (Ref. 11)

$$V_D = -\left(\frac{kT_e}{2e} \right) \ln \left(\frac{T_e m_i}{T_i m_e} \right) \tag{10}$$

where m_e and m_i are electron and ion mass, respectively. For the case of the hydrogen molecular ions, V_D is approximately 20 volts, when T_e is about 35000 K. Similarly, it can be shown

(Ref. 9) that the potential difference V_b between the axis and the boundary sheath is

$$- V_b \simeq \frac{kT_e}{e} \ln (R/1.7\,\lambda_e)$$

where λ_e is the mean free path of the electrons. So, for the case of hydrogen gas dissociator, the ions created at the axis are accelerated to the wall through a potential $V_T = V_b + V_D$ with $V_T = 26$ V.

We are now in a position to determine the efficiency of the source. We can write a diffusion equation similar to Eq. (2) for the case of hydrogen atoms as produced by electron impact on the molecules:

$$\phi_H = D \nabla n_H \tag{11a}$$

Here ϕ_H is the flux of H atoms, D the diffusion constant for the diffusion of atoms through H_2 molecules, and n_H the number density of atoms. Since each electron impact resulting in the dissociation of a molecule produces two hydrogen atoms, taking the divergence of (11a) yields:

$$\nabla \cdot \phi_H = 2\nu_H n \tag{11b}$$

where ν_H is the collision frequency of electrons with molecules resulting in dissociation and n is the electron density. Equation (11b) implicitly contains the fact that the density of electrons is much smaller than the density of hydrogen molecules. We now combine Eqs. (11a) and (11b) to get:

$$D \nabla^2 n_H = n \nu_H \tag{12}$$

Similarly from Eq. (2):

$$D_a \nabla^2 n = n \nu \tag{13}$$

Solving for n in Eqs. (12) and (13), we get

$$\frac{D_a \nabla^2 n}{\nu} = \frac{D \nabla^2 n_H}{\nu_H}$$

or

$$\nabla^2 n_H = \frac{2\nu_H}{\nu} \frac{D_a}{D} \nabla^2 n \, ,$$

resulting in

$$D \nabla n_H = \frac{2\nu_H}{\nu} D_a \nabla_n$$

or finally

$$\phi_H = \frac{2\nu_H}{\nu} \phi \tag{14}$$

Now, the rate at which hydrogen atoms arrive at the wall is $n_H V/4$, where V is the thermal velocity of the atoms. A fraction γ of the atoms arriving at the wall then recombine to form hydrogen molecules. In order to achieve equilibrium, the rate of production of atoms and the rate of their loss have to be equal,

$$\gamma n_H \frac{V}{4} = \frac{2\nu_H}{\nu} \phi \; . \tag{15}$$

At this point we can relate the ion flux ϕ to the power input into the source. If we assume that the only loss mechanism in the source is due to the energy carried away by the ions falling to the wall, then we will have, at equilibrium, $P = V_T I$, where P is the power input into the source and I is the ion current. Since we have $e\phi = j$, where j is the current density, we can use Eq. (15) to obtain

$$\gamma n_H \frac{V}{4} = \frac{2\nu_H}{\nu} \frac{j}{e}$$

or using the relation for power

$$\gamma n_H \frac{V}{4} = \frac{2\nu_H}{\nu} \frac{P}{AeV_T}$$

where A is the area of the source. For a source with efficiency of atom production η, we have $n_H = 2\eta n_0$, where n_0 is the density of molecules. Combining this with the last equation we obtain an expression for the source efficiency η:

$$\eta = \frac{4\nu_H}{e\nu} \frac{P}{V\gamma n_0 A V_T}$$

or:

$$\eta = \frac{4\nu_H}{\nu} \frac{kT_H}{e} \frac{P}{AV\gamma V_T p} \tag{16}$$

If we assume $\gamma = 5 \times 10^{-5}$ (Ref. 12), $p = 0.1$ torr $= 7.49 \times 10^{-5}$ dynes/cm^2, and $P = 10$ W, the efficiency of a source with a radius of 1 inch and length of 2 inches is of the order of 5 percent, in general agreement with experimental results.

The assumptions leading to Eq. (16) for efficiency preclude the applicability of the expression for η in the regime of high pressures, where certain concepts such as the sheath potential V_D are likely to fail. The assumption of absence of negative ions (which promote volume recombination) and neglect of other loss mechanisms also limit the accuracy of η. Finally, one should bear in mind that an increase in power results in the increase of T_e, which affects most of the parameters in the expression for η.

V. Particle Interactions With Dissociator Walls

The interaction of particles in the hydrogen gas dissociator with dissociator walls is an important effect which directly influences both the atom flux from the source and the source life (Refs. 12, 13). Various ions and neutral particles in the source which drift onto walls interact in different ways with the glass walls of the dissociator. Hydrogen atoms form a layer on the wall by an adsorption mechanism which can subsequently promote the formation of molecules. It is also known that the silica glass can be permeable to H_2 molecules with very high energies. Various chemical interactions between the particles and the molecules in the glass will also be important in recombination of atoms and introduction of impurities in the plasma.

Equation (10) expresses the relationship between V_D and the electron temperature T_e. It is then clear that an increase in the electron temperature increases the energy with which ions strike the walls. The resulting ion sputtered surface is detrimental to the efficiency η, presumably because of an increase in the microscopic surface area generated by surface granules and the subsequent increase in recombination rate. Striking ions also may release other ions from the wall material, including impurities in the glass. This results in the absorption of power by the new ions and decrease in hydrogen atom production efficiency. The same consideration applies equally well to the presence of water vapor or other impurities in the source. Finally, carbon atoms present as impurities in the vacuum system may deposit on the surface and directly influence the value of surface recombination rate γ (Ref. 12).

VI. Summary, Conclusions and Recommendations

We have examined some of the processes which are important in the study of the hydrogen gas dissociator. We have seen that collisional processes in the dissociator result in the production of hydrogen atoms and other particles, as well as photons. By approximating the RF discharge with the positive column of a dc plasma source, we considered the transport properties of the plasma to obtain relationships for electron temperature and source efficiency in terms of gas pressure, input power, and geometric size of the source. Finally, we briefly outlined the influence of the wall of the dissociator on its performance.

On the basis of this study, the following conclusions regarding the design of more efficient RF hydrogen atom sources may be inferred. Since the wall recombination presents the greatest limitation at the source, care should be taken to construct the source from materials with the lowest recombination coefficient. At the present time, Pyrex is the most widely used material for this purpose. It is recommended that other materials with smaller coefficient be sought for possible application in source construction. Furthermore, sources should be acid cleaned not only to remove any surface deposits that may interfere with the development of an optimized plasma temperature, but also to leave a smooth surface to minimize the area which can enhance atom recombination at the walls. Since the wall recombination coefficient is a strong function of temperature (Ref. 12), designs should include adequate measures for the removal of the heat generated by the RF power in the dissociator bulb. While the considerations in this study were based on RF power inductively coupled to the source, it is believed that they apply equally well to the case of capacitive coupling. In either case, care should be taken to couple the RF power into the source with adequate efficiency.

Relations (9) and (16) may be used in both design and operation of the source. As indicated by these equations, the physical dimension of the dissociator strongly affects its performance. However, it should be carefully noted that while an increase in the size of the dissociator decreases the sputtering effect, it can also decrease the efficiency if power and pressure are held constant. It therefore appears that a careful design for a dissociator may employ relations (9) and (16) to seek an optimized size for the source and find a balance between these two effects. In general, however, it can be said that for constant power and pressure, a smaller dissociator leads to more efficiency, while a larger size results in longer lifetime.

Equations (9) and (16) can also be used to optimize the performance of a dissociator by properly adjusting the pressure and power. It should, however, be kept in mind that these relations do not hold for the case of high pressures (in excess of 500 microns). Finally, it should be reemphasized that the evaluation of the source performance by examination of the color of the plasma is inadequate. Barometric methods are more direct and reliable and should be preferred.

Future studies of the RF hydrogen dissociator should include a search for wall materials with reduced recombination coefficient. It is also recommended that considerations be given to the application of an axial magnetic field on the source to confine the ions close to axis and reduce the ion bombardment of the walls, similar to RF ion sources. If a magnetic field can be applied on the source without appreciable degradation of the efficiency, it may be possible to increase the life of the source by an appreciable amount.

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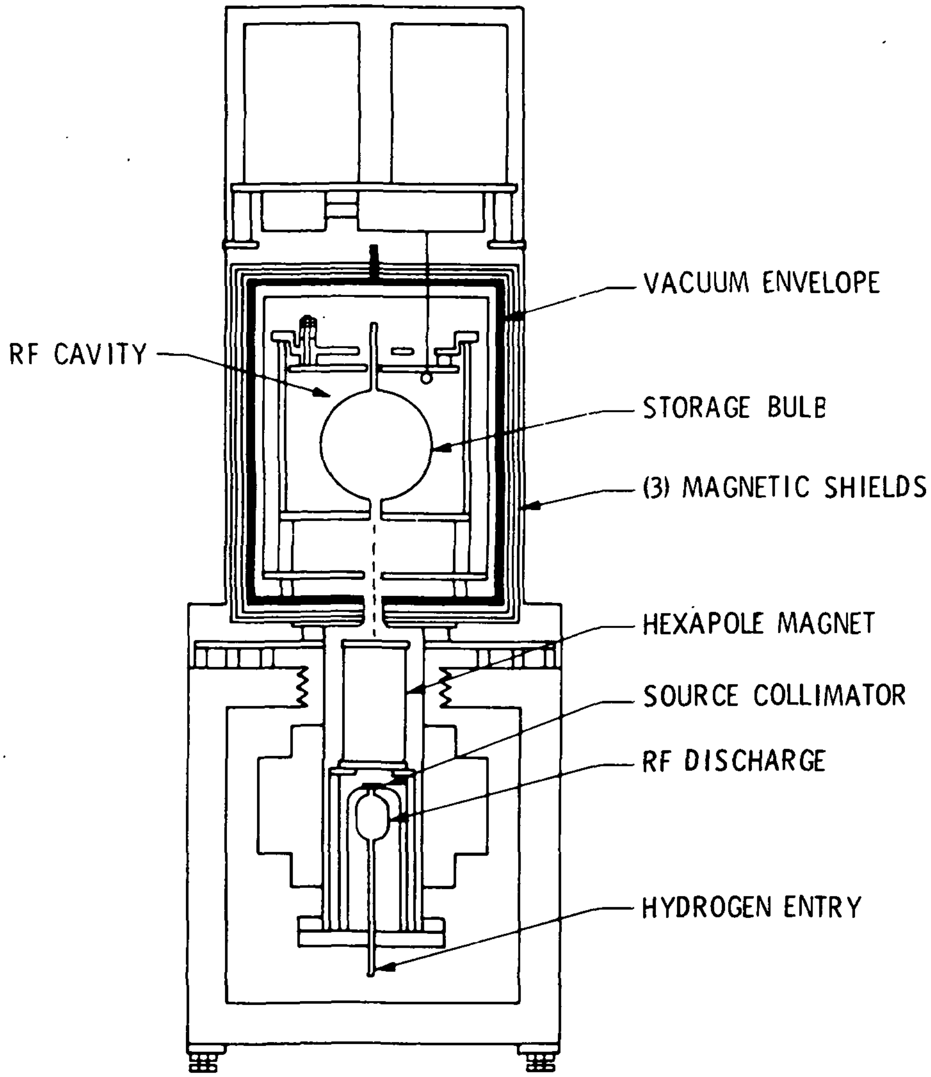

Fig. 1. Line drawing of the hydrogen maser