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RECOMBINATION OF IONS AND ELECTRONS IN A HIGHLY IONIZED HYDROGEN PLASMA

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William S. Cooper III and Wulf B. Kunkel

October 1, 1964

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ABSTRACT

A hydrogenic plasma with an electron density near 5×10¹⁵ cm⁻³ and a temperature between 1 and 3 volts is produced by a powerful transient discharge in a tube of 14 cm diameter and 86 cm length. Spectroscopic techniques are used to follow the subsequent decay of both electron density and temperature for about 300 µsec. A strong uniform axial magnetic field suppresses rapid radial particle losses, but does not appreciably reduce the radial energy transport. The axial transport apparently results merely in the slow growth of relatively thin dense boundary layers at the tube ends. In the interior the ionization is shown to decay primarily by volume recombination, and the rate coefficient agrees well with that calculated by Bates, Kingston, and McWhirter for plasmas that are virtually opaque to the Lyman-line radiation. Evidently, the plasma starts out essentially fully ionized and seems to remain close to local thermal (Saha) equilibrium for 120 µsec. The inferred energy loss rate, on the other hand, initially exceeds that expected for kinetic transport by perhaps an order of magnitude, indicating that early in the afterglow radiative transfer may be significant.

I. INTRODUCTION

Several authors, beginning with Lord Rayleigh in 1944, have observed recombination in dense hydrogen plasmas. 1-6 The recombination was at first attributed to "radiative recombination," described by

$$H^{+} + e \rightarrow H^{*} + h\nu, \tag{1}$$

with an associated recombination coefficient a defined by

$$\frac{dN_i}{dt} = -\alpha(T)N_iN_e, \qquad (2)$$

where N_i is the ion density, N_e is the electron density, and T is the temperature. Measured values of a have been too large, sometimes by more than a factor of 10, to be explained by radiative recombination alone. D' Angelo showed that another process, "three-body recombination," could contribute significantly to the recombination rate of a dense plasma. This process is simply the inverse of electron collisional ionization:

$$H^{+} + e + e \rightarrow H^{*} + e.$$
 (3)

Several recent publications by Bates and Kingston, ^{8,9} McWhirter, ¹⁰
Bates et al., ^{11,12} Stubbs et al., ¹³ Byron et al., ¹⁴ and Hinnov and Hirschberg ¹⁵
have dealt with theoretical aspects of the recombination process. Hinnov and Hirschberg, Kuckes et al., ¹⁶ Motley and Kuckes, ¹⁷ and Robben et al. ¹⁸ also give experimental results, determined from measurements of medium-density (N_e ≤ 5×10¹³ cm⁻³) low-temperature (T ≤ 3000 °K) hydrogen or helium plasmas. The most complete theoretical results are due to Bates, Kingston, and McWhirter, who have calculated both recombination rates and distributions of excited states in recombining hydrogen plasmas by writing down the rate equations of all processes populating and depopulating the excited states and the ground state, using known or assumed cross sections, and solving them simultaneously with the aid of a computer. ^{9,11,12} They introduce a "collisional-radiative decay coefficient" γ_{1,1} defined analogously to Eq. (2) by

$$\frac{dN_1}{dt} = \gamma(N_1, N_e, T_e)N_iN_e, \qquad (4)$$

where N₄ is the density of hydrogen atoms in the ground state.

In an electron-proton plasma we have, of course, $N_i = N_p$; and if $kT_a < < eV_4 = 10.2 eV$, practically all neutral atoms are in the ground state, so that N₄ + N₂ = const is a very good approximation. In spite of these simplifications, Eq. (4) alone is still insufficient to predict the decay of an actual recombining plasma because the electron temperature To(t) needs to be determined in a selfconsistent manner. In general the behavior of T (t) follows from considerations of energy flow into and out of the electron gas. These considerations have been included in an approximate manner in some of the literature cited, 14, 16, 17 and rather detailed calculations of the energy balance in certain decaying plasmas have recently been completed by Bates and Kingston. 19, 20 The analyses invariably make use of the fact that the electrons in decaying plasmas transfer energy only locally to the ions and atoms. The problem is kept tractable by the further simplifying assumption that the temperature either of the atoms alone or of both the ions and the atoms is somehow maintained in a fixed ratio to the wall temperature. The first of these models is applicable to the magnetically confined afterglows in Stellarators at low gas densities. Such approximations are not justified, however, when the plasma density is so high that the temperature of the atoms remains closely coupled to that of the electrons.

In this paper we are interested in the nature of the decay of a highly ionized hydrogen plasma at densities above 10¹⁵ cm⁻³ and temperatures in the neighborhood of 10 000 °K. Spectroscopic techniques were used to determine both temperature and electron density as a function of time and of position in a cylindrical plasma of 14.6 cm diameter and 86 cm length embedded in an axial magnetic field of 16 kG

(see Fig. 1). The details of the measurements and most of the observed results are reported elsewhere. ²¹ Here we summarize only those facts that are pertinent to the discussion of the decay process.

II. PLASMA CONDITIONS

The plasma is prepared by a pulsed high-current discharge in hydrogen gas at 0.1 torr initial pressure. A few microseconds after the power supply has been shortcircuited all potentials between the electrodes are zero and the plasma is allowed to cool off. This technique has been described before, ^{22,23} and we merely list the parameters of the resulting plasma in Table I.

The density of neutral atoms has not been measured directly, but may be inferred in retrospect (see Section 3). It is clear, however, that the plasma starts out highly ionized and for most purposes can be considered as collision-dominated from the beginning. It is inconceivable that the ion temperature differs much from that of the electrons, and the electrons must have a velocity distribution that is very nearly Maxwellian. Because of the large cross section for resonant charge transfer, the neutral atomic component may also be considered as tightly coupled to the plasma, so that the entire mixture can be adequately described as a single fluid with well-defined macroscopic properties. In particular, it turns out that the recombination rate is much larger than the rate of interdiffusion of atoms and ions, so that the drift of these two species with respect to each other may be neglected as a macroscopic motion. However, as is well known, the energy transport associated with these drifts has to be included as a contribution to the generalized heat-flow vector. ²⁴

The latter considerations permit an estimate of the growth rate of the thermal boundary layers that must exist between the plasma and the end plates of the tube. 25 The gas in these boundary layers must be relatively dense, since no

pressure gradients can persist in a direction parallel to the magnetic field. Substitution of reasonable values for the thermal diffusivity then shows that during the time of the experiment the thickness of the layers should grow only to about 1 cm or 2. The spectroscopic observations indeed showed that the plasma was reasonably uniform along the axis, at least to within 2 cm from the end plates. 21 The plasma is evidently not decaying primarily by transport along the magnetic field lines.

The situation is quite different in regard to radial gradients. Here the pressure balance is easily dominated by the magnetic field, and no dense boundary layer is required. In the absence of flute instabilities or anomalous diffusion effects the radial drift velocity of this plasma cannot exceed a few meters per second, and, indeed, the experimental measurements gave no indication of any significant cross-field diffusion. The collisional energy transport, on the other hand, is not much affected by the magnetic field under the existing conditions. Hence, when the dense boundary layer is lacking, the radial temperature gradients may easily extend over several centimeters, i.e., over a large fraction of the tube diameter, just as observed in this and similar experiments. We therefore arrive at the conclusion that the plasma does not decay by either radial or longitudinal loss of charged particles, but primarily by radial energy transport accompanied by recombination in the volume.

III. COLLISIONAL-RADIATIVE RECOMBINATION

As mentioned before, both the electron density and the temperature of the plasma have been measured in these experiments. It is therefore possible to compare the results directly with the calculations by Bates and co-workers without reference to the nature of the energy flow. We merely require that the disappearance rate of the ions and electrons be controlled by local conditions and do not depend directly on the existing gradients. A very carefully prepared set of data, obtained at a radial position 3.35 cm from the tube axis, is presented in Figs. 2 and 3. It is seen that at early times the temperature drops rapidly while the density changes only little. It should be noted that the estimated errors do not take into consideration the nonuniformities along the line of sight that exist during the first 50 or 60 µsec. After about 80 µsec the measurements should merit comparison with theory, however.

Since during the entire decay period almost all the neutrals are in the ground state, we may assume $\hat{N}_1 = -\hat{N}_i$, permitting us to measure the decay coefficient γ , defined in Eq. (4), from the observed values of N_i and \hat{N}_i . We have calculated γ as a function of time for the decay of the plasma shown in Fig. 3. These values are presented in Fig. 4 together with theoretical values of γ calculated from the work of Bates et al., $^{11, 12}$ using the observed values of the electron density and temperature at each time. It should be emphasized that we are dealing with simultaneous measurements of the electron density and temperature on a single shot, and so are not depending upon shot-to-shot reproducibility. Other shots, however, showed a similar behavior. The theoretical values of γ are shown for two cases: in the first, the plasma is assumed to be optically thin (Curve A); in the second, the plasma is assumed to be opaque to the lines of the Lyman series (Curve B).

The values of γ early in the decay period depend also on N_4 , the density of neutral hydrogen atoms in the ground state, because the electron temperature is still high enough to cause significant reionization and reexcitation from the ground state. In calculating theoretical values of γ we have evaluated N_4 by assuming that (a) the plasma is initially fully ionized, enabling us to equate the total particle density to the ion density early in time, and (b) the total particle density in the region of plasma observed remains constant during the decay period, enabling us to calculate N_4 by subtracting the measured ion density from the total particle density. The first assumption we justify in retrospect; the second seems very reasonable, as the estimated times for diffusion are much longer than the observed decay times.

The plasma is certainly not completely opaque to the entire Lyman series, as was assumed in the theoretical treatment leading to Curve B, as the lines are broadened by Stark and Doppler broadening, and the light in the wings of the lines must escape the plasma with little reabsorption. This escape of radiation will tend to increase y, as it essentially represents a new mechanism for populating the ground state. However, with this plasma it is easy to show that for the L line the Doppler broadening is always much greater than the Stark broadening, and furthermore that the plasma is opaque to L during the entire decay period. Of all discrete transitions populating and depopulating the ground state the term corresponding to emission and absorption of L is by far the largest. We therefore expect that even if L_a is the <u>only</u> line in the Lyman series that is reabsorbed, the assumption that all the Lyman lines are reabsorbed will yield a reasonable estimate of the decay coefficient. Measurements of y in this decaying plasma should therefore fall between the two theoretical curves in Fig. 4, but probably should be closer to Curve B. This is seen to be the case. Besides, the calculations by Bates et al. make use of approximate cross sections, and it is not clear that the theoretical values are reliable to within less than a factor of 2.

It is instructive to plot the ion density versus temperature in a single "decay curve" or "cooling curve," shown in Fig. 5. For the first 40 µsec the temperature drops but the ion density remains constant, as would be expected if the plasma were initially completely ionized. Also shown in Fig. 5 is an equilibrium cooling curve calculated from Saha's equation (labeled "LTE") and fitted to this early portion of the decay curve, assuming a total proton density of 4.6×10¹⁵ cm⁻³. The error boxes show the estimated experimental errors; as already mentioned, the points before 80 µsec are more uncertain than indicated because of the longitudinal nonuniformities. Apparently the plasma at this radius is initially fully ionized. This is to be expected, because even if the temperature early in the decay were overestimated by as much as 50% according to the calculations by Bates et al. the degree of ionization would be very high even in the steady state, and must of course be still higher in a decaying plasma. It is to be noted that the initial ion density is less than the original hydrogen-atom density of 5.8×10¹⁵ cm⁻³ (indicated by the short solid line). This is not really surprising, since we have already pointed out that the plasma must be bounded by boundary layers of considerably higher density, particularly near the two end plates. In addition, violent motion is produced during the plasma-forming discharge, and therefore there is no real reason to expect a original density to exist when the plasma is finally immobilized by the magnetic field.

Figure 5 also indicates that the ionization remains quite close to LTE during the first 120 µsec and then departs noticeably from the Saha equilibrium. A discussion of this behavior requires information concerning the energy transport which, in the final analysis, controls the plasma decay.

IV. ENERGY DECAY

In the absence of both mass flow (convection) and electric currents (ohmic heating) the conservation of energy can be expressed in the simple form

$$\frac{\partial \mathcal{E}}{\partial t} + \nabla \cdot \underline{Q} = R . \tag{5}$$

The symbol & represents the energy density, Q the particle-energy flow vector, and R the net absorption rate per unit volume of radiant energy. In the case discussed here we can ignore the energy of molecular dissociation, because the hydrogen remains atomic throughout the period of interest. We can also neglect the energy stored at any given instant in the form of electronic excitation. Thus we have as a good approximation

$$\mathbf{\xi} = \epsilon_i N_e + \frac{3}{2} (N_0 + N_e) kT, \qquad (6)$$

where ϵ_i = 13.6 eV is the ionization energy per atom. N_0 is the initial proton density, hence it is constant in time, but in our case N_0 may be a function of radial position. From Figs. 2 and 3 it is clear that, after 50 µsec, $\epsilon_i \partial N_e / \partial t$ constitutes the major portion of the energy decay rate.

It is not obvious that the energy-flow vector Q for reacting gas mixtures such as a partially ionized gas can always be expressed in the form $Q = -K(T) \nabla T$, because the transport is in general strongly affected by the interdiffusion of the various components. The situation is much simplified in our experiment, however, since we have already shown that the energy transport is predominantly radial and that radial diffusion of plasma in this case is adequately suppressed by the strong axial magnetic field. We therefore should be able to write Eq. (5) as

$$(\epsilon_i + \frac{3}{2}kT)\frac{\partial N_e}{\partial t} + \frac{3}{2}(N_0 + N_e)k\frac{\partial T}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}rK(r,t)\frac{\partial T}{\partial r} + R(r,t), \quad (7)$$

where K(r,t) is the local ordinary coefficient of thermal conductivity of our hydrogen plasma.

When the gas is very close to LTE the composition is a function of temperature and pressure (or density) only and therefore $K = K(T, N_0)$. Curiously, no measurements of K for high-temperature partially ionized hydrogen are as yet available in the literature. We may make a crude estimate based on a simplified kinetic theory model, of course, $\frac{29}{100}$

$$K \approx \sum_{j} K_{j} = \frac{75k}{256} (8\pi kT)^{1/2} \sum_{j} N_{j} \left[\sum_{\ell} \sigma_{j\ell} N_{\ell} \left(\frac{m_{j} m_{\ell}}{m_{j} + m_{\ell}} \right)^{1/2} \right]^{-1}$$

(8)

where the summations extend over the three species atoms, protons, and electrons, identified by the subscripts a,i, and e respectively. The masses and effective collision cross sections are denoted by the conventional m_j and $\sigma_{j\ell}$. Since the number densities N_j are shown explicitly in Eq. (8), the expression can be used even when the composition deviates from that given by Saha's equation.

When the contribution by the free electrons is significant the value of K_e is presumably very close to that for a fully ionized gas. The contribution from K_i is always negligible and the only quantity in question is K_a . Using as first guesses $\sigma_{aa} = 10^{-15} \text{ cm}^2$ and $\sigma_{ia} = 5 \times 10^{-15} \text{ cm}^2$ (resonant charge exchange 31), and substituting observed values of N_i and T_i , we can compute K(r,t) for our plasma. The result is shown in Fig. 6 for r=5 cm, plotted against temperature

rather than time. This curve does not differ appreciably from more refined calculations in which equilibrium ionization is assumed. 32 K_a is a decreasing function of temperature for $T > 5000\,^{\circ}$ K because N_a/N_e decreases. K_e is smaller than K_a below 10000 °K because the Coulomb collision cross section becomes very large. In Fig. 6 we have overestimated K_e for our cooling problem because we have ignored the effect of the magnetic field. The latter effect is probably offset in part by thermoelectric currents which must be circulating in finite plasmas with gradients such as ours. A rigorous analysis of the heat-flow problem would be very involved indeed.

The radiative contribution R(r,t) is even more difficult to estimate because the emissivity and absorption coefficient are very rapidly varying, complicated functions not only of position and time but also of frequency, particularly in the region of the Lyman spectrum. It is very important to consider the absorption carefully because the power density emitted, for example, in the L_a line alone, $J_a = eV_1A_{24}N_2$, is readily shown to be near 3×10^9 erg/cm³-sec in most of the plasma, and this is almost ten times the entire value of $\partial \xi/\partial t$ at r = 3.35 cm. On the other hand, the absorption coefficient for the Doppler-broadened L_a is given by $\frac{38}{2}$

$$k_{12} = 6 \times 10^{-12} N_1 T^{-1/2} cm^{-1}$$
, (9)

where T is again in degrees Kelvin and N₁ is the number of ground-state atoms per cm³. Thus during most of the decay k₁₂ is quite large, in agreement with the findings of Fig. 4, and the resulting contribution of J_a to R should be rather small. It is not clear, however, that this argument permits us to neglect R(r,t) when the radiative transfer is integrated over all frequencies. Radiation from impurities may also contribute significantly. In fact, the following observation may be taken as an indication that radiation plays an important part in the cooling process of the decaying plasma described here.

Measurements of N_e(t) and T(t) like those shown in Figs. 2 and 3 have been performed for different radial positions. ²⁴ It is thus possible to solve Eq. (7) for an upper bound of K(r,t) by setting R = 0 and integrating numerically over a suitable radial interval. The results of this procedure are included in Fig. 6. These values are estimated to be reliable only to within a factor of 2 as indicated, because the temperature gradient could not be evaluated with a higher precision. It is seen that the energy decay rate is consistently substantially higher than expected from kinetic thermal conductivity, particularly at the higher temperatures, i.e., at the higher degrees of ionization. This is the region where the plasma tends to be less opaque. Unfortunately, the experiment did not permit a direct measurement of the far ultraviolet radiation escaping from the plasma.

V. CONCLUSIONS

In the foregoing discussion we have attempted to demonstrate that the decay of a magnetically confined highly ionized plasma is governed by the energy removal rate. It appears that the hydrogen plasma investigated experimentally cools more rapidly than can be expected from kinetic heat transfer, presumably because of contributions from radiative heat transfer. This occurs in spite of the fact that during most of the decay the plasma is expected to be nearly opaque to the Lyman line radiation. The observed decay coefficient agrees rather well with that predicted by Bates and co-workers. During the first 120 µsec the degree of ionization in the interior of the decaying plasma remains close to LTE, i.e., we have

$$\frac{\partial N_{e}}{\partial t} \approx \left(\frac{\partial N_{e}}{\partial T}\right)_{Saha} \cdot \frac{\partial T}{\partial t} . \tag{9}$$

However, no special significance must be attached to this feature. It merely means that in this region the energy removal rate is slow enough for the ionization to remain temporarily in a quasi-steady state, and in this case the latter happens to be indistinguishable from the Saha equilibrium. 12

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FOOTNOTES AND REFERENCES

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Table I. Parameters of decaying hydrogen plasma.

Quantity	Symbol	Value
Length	L	86 cm
Diameter	2R	14.6 cm
Magnetic field	В	16 kG
Initial pressure	P ₀	0.10 torr
Electron density	Ne	4.5→0.5×10 ¹⁵ cm ⁻³
Plasma temperature	T 8.00	2-0.8×10 ⁴ °K
Decay-time constant	7	≈ 10 ⁻⁴ sec
Electron gyrofrequency	ω ce	3×10 ¹¹ sec ⁻¹
Ion gyrofrequency	ω _{ci}	1.5×10 ⁸ sec ⁻¹
Electron-ion collision frequency	ν ei	7-25×10 ¹⁰ sec ⁻¹
Electron-ion equilibration time	τ _{eq}	≈ 10 ⁻⁷ sec
Plasma magnetic energy-density ratio	β	< 3×10 ⁻³
Estimated atom density	Na	0→4×10 ¹⁵ cm ⁻³
Inferred atomic charge-exchange frequency	vexch	25-3×10 ⁶ sec ⁻¹

FIGURE CAPTIONS

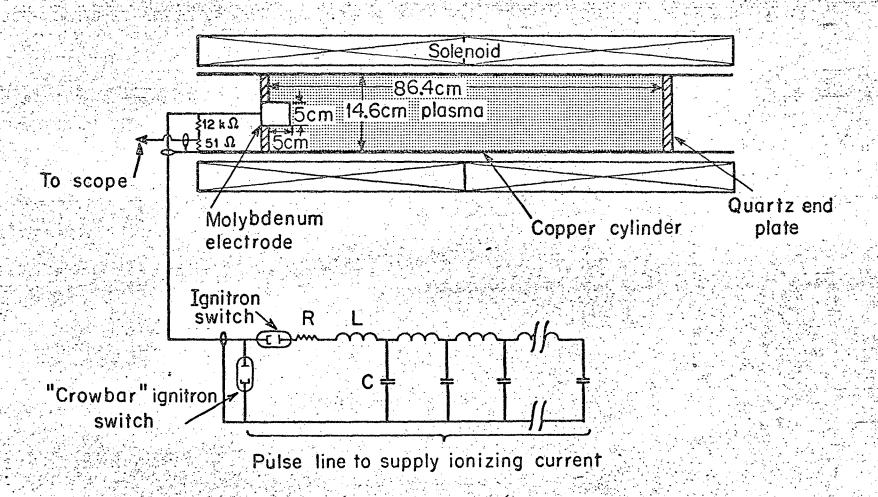
- Fig. 1. Schematic diagram of apparatus used to produce plasma.
- Fig. 2. Weighted mean value of the ion density on a typical shot as a function of time at a radius of 3.35 cm. Estimated errors are shown.
- Fig. 3. Weighted mean value of the temperature on the same shot as Fig. 2 as a function of time at a radius of 3.35 cm. Estimated errors are shown.
- Fig. 4. Typical values of the decay coefficient γ as a function of time. Curves

 A and B show theoretical values of γ for a transparent plasma and for a

 plasma opaque to the Lyman lines, respectively. These curves may be

 subject to a common systematic error of a factor 2 or 3. Estimated

 experimental errors are shown for the measured values of γ.
- Fig. 5. Decay curve for the shot shown in Figs. 2 and 3. The solid curve labeled LTE was calculated for Saha's equation and was fitted to the early portion of the decay curve.
- Fig. 6. Thermal conductivity computed from Eq. (8) (solid curve) and observed coefficient inferred from Eq. (7) if R(r,t) were negligible.



IFig. 1

MU-22165-A

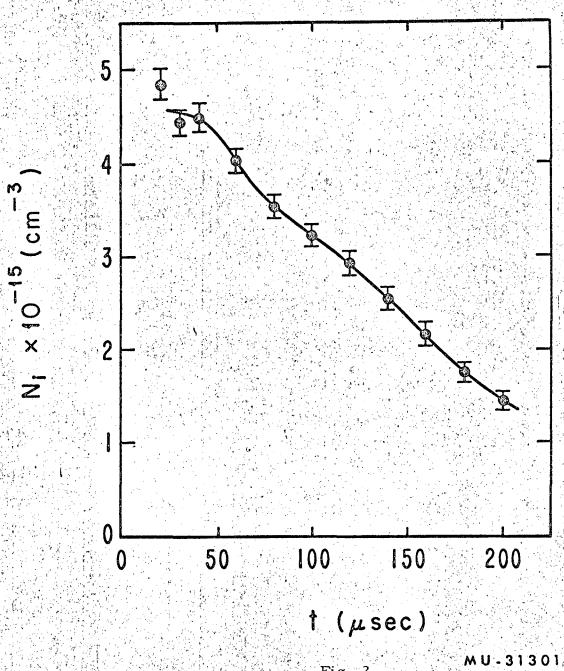
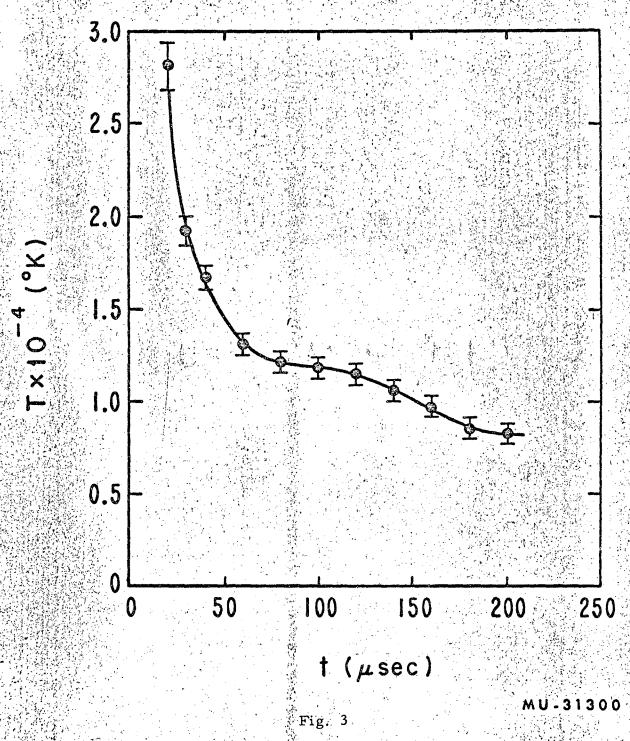


Fig. 2



1800

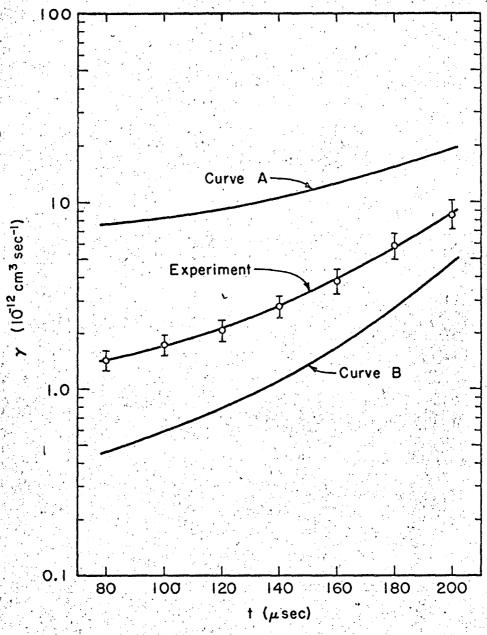


Fig. 4

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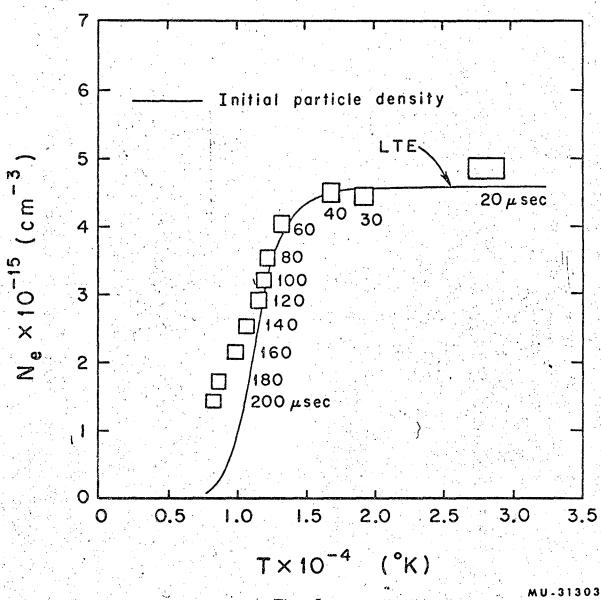


Fig. 5

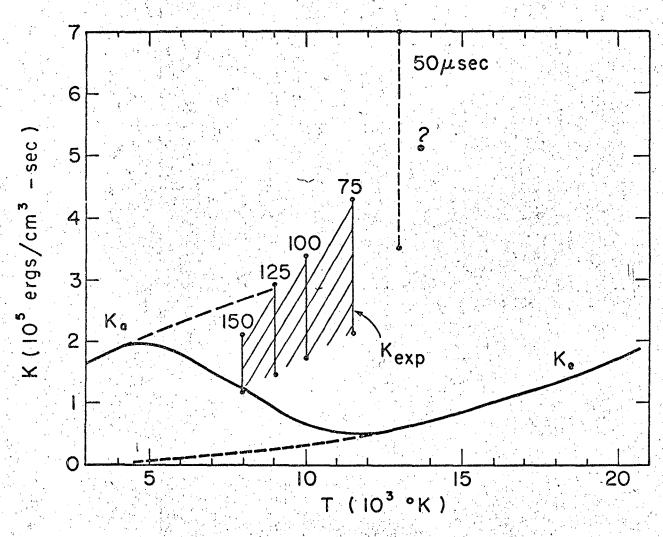


Fig. 6 MUB-4354

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