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The Decay of Optically Thick Helium Plasmas, Taking into Account Ionizing Collisions between Metastable Atoms or Molecules

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THE DECAY OF OPTICALLY THICK HELIUM PLASMAS, TAKING INTO ACCOUNT IONIZING COLLISIONS BETWEEN METASTABLE ATOMS OR MOLECULES

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ABSTRACT

The effective recombination rate of a helium afterglow plasma, which is optically thick towards the resonance lines, is calculated from the coupled rate equations for the number densities of free electrons and of metastable atoms or molecules. The model employed is a neutral plasma, consisting of one kind of ions and one kind of metastables. The ions are lost by electron-ion recombination only, with subsequent formation of metastables, which are then deactivated in collisions with free electrons or with other metastables: in the latter case one electron is regained to the free state. When the rate constants for these various processes are time-independent, it is found that after a certain transition time a transient equilibrium between the number densities of electrons and metastables is attained. In a dense afterglow plasma, where the recombination coefficient may be large, the transient equilibrium density of metastables may become significantly higher than the quasi-equilibrium value obtained by equating the time derivative of the metastable density to zero, and the effective recombination coefficient may be reduced by much more than a factor of two.

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1. INTRODUCTION

A large amount of experimental work on electron-ion recombination in helium discharge plasmas has shown that, under current afterglow conditions, the plasma disintegrates in reasonable agreement with the model of collisional-radiative recombination. This model has been worked out in articles by Bates et al. [1], Bates and Khare [2], Bates et al. [3], and Collins [4].

In the theoretical approach of Bates et al. [1] a rate equation is written for each excited level n of the neutral atom, describing the rate of increase of the population density N_n with time. Thus an infinite set of coupled differential equations is obtained, which yields the course of the recombination. Except for extremely dense plasmas, the population densities N_n $(n \neq 1)$ are much smaller than the density N of free electrons, and so the same is valid for the time derivatives of N_n and N. This means that a quasi-equilibrium number density is established almost instantaneously for the excited levels, and the time derivatives $\dot{N}_n$ $(n \neq 1)$ occurring in the set of differential equations can be put equal to zero, which greatly simplifies the solution.

In a later paper by Bates et al. [3] the influence of ionizing collisions between two metastable atoms on the overall recombination rate was taken into account. This calculation was also based on an assumption of quasi-equilibrium number density of metastables, and it was shown that the effective recombination coefficient may then be reduced by at most a factor of two. However, as was pointed out already in the former paper [1], in a plasma which is optically thick to the resonance lines the density of atoms in the quantum level $n = 2$ may become comparable to the free electron density, and the assumption of quasi-equilibrium metastable density is then no longer justified. This is a common situation in an afterglow plasma, and the

calculation of the metastable density and effective recombination coefficient in such a plasma will be the subject of the present paper.

2. PHYSICAL MODEL

We assume a two-level model of the helium atom or molecule: a ground level with quantum number n = 1, which for the helium molecule is repulsive, and an excited, metastable level with quantum number n = 2. In addition, we consider the unbound level n = ∞. The plasma is neutral, that is, the free electron density is equal to the number density of ions; further, there is just one kind of ions and one kind of metastables (atomic or molecular). The free electrons recombine with the ions in an unspecified way to form a metastable state, and the rate constant for this process is called the recombination coefficient α. The metastables are deexcited to the ground level in collisions with free electrons with a rate constant β. Finally, two metastables may collide with each other, such that one is deexcited and the other becomes ionized. The rate coefficient for this process is denoted γ. All other processes, such as ambipolar diffusion, are assumed to be negligibly slow. Further, in a first consideration, the three rate constants α, β, and γ are assumed to be time-independent.

This simplified model might be applied to several limiting cases. First, when the dominant ion is the atomic He^+, the collisional-radiative recombination results in the formation of atomic metastables He(2), which undergo mutual ionizing collisions to again produce atomic ions:

$$He^+ + e + X \xrightarrow{\alpha} He(2) + X \qquad (1)$$

$$He(2) + He(2) \xrightarrow{\gamma} He(1) + He^+ + e \qquad (2)$$

Here, the stabilizing particle X may be an electron or a neutral

atom. If, on the other hand, only molecular ions He_2^+ are present in the afterglow, collisional-radiative recombination results in the formation of molecular metastables $He_2(2)$, which, colliding with each other, produce atomic ions, as was demonstrated by Collins and Hurt [5]. If the pressure is high enough, the atomic ions are immediately converted into molecular ions:

$$He_2^+ + e + X \xrightarrow{\alpha} He_2(2) + X \tag{3}$$

$$He_2(2) + He_2(2) \xrightarrow{\gamma} 3He(1) + He^+ + e \tag{4}$$

$$He^+ + 2\,He(1) \longrightarrow He_2^+ + He(1) \tag{5}$$

Finally, the dominant ion may be He_2^+, undergoing dissociative recombination to form atomic metastables He(2):

$$He_2^+ + e \xrightarrow{\alpha} He(1) + He(2) \tag{6}$$

These metastables may obey the process (2), forming atomic ions, which again, if the pressure is high, are rapidly converted according to (5). In this case we have molecular ions but atomic metastables, and the recombination radiation observed will be atomic line emission.

Denoting the metastable density by M, the free electron density by N, and the time derivatives by $\dot{M}$ and $\dot{N}$, this physical model leads to the coupled rate equations

$$\dot{M} = \alpha N^2 - \beta MN - 2\gamma M^2 \tag{7}$$

$$\dot{N} = -\alpha N^2 + \gamma M^2 \tag{8}$$

If M is substituted against the ratio p = M/N, the rate equations become

$$\dot{p} = -N[\gamma p^3 + 2\gamma p^2 - (\alpha - \beta)p - \alpha] \tag{9}$$

$$\dot{N} = -N^2(\alpha - \gamma p^2) \tag{10}$$

From eq. (10) it is seen that the effective recombination coefficient is given by the expression

$$\alpha_{eff} = \alpha - \gamma p^2 \tag{11}$$

3. SOLUTION OF DIFFERENTIAL EQUATION FOR THE RATIO p

Next we shall determine the time development of the different particle concentrations. It is then simplest to study the ratio p between the metastable density M and the free electron density N. The rate equations (9) and (10) can be combined into one second-order differential equation for the density ratio p:

$$\ddot{p} = \frac{2\gamma p^2 + 4\gamma p + \beta}{\gamma p^3 + 2\gamma p^2 - (\alpha - \beta)p - \alpha} \dot{p}^2 \tag{12}$$

After one integration, equation (12) yields

$$\dot{p} = C\,|p - p_1|^{A_1}\,|p - p_2|^{A_2}\,|p - p_3|^{A_3} \tag{13}$$

p_1, p_2, and p_3 are the solutions of the equation

$$\gamma p^3 + 2\gamma p^2 - (\alpha - \beta)p - \alpha = 0 \tag{14}$$

and A_1, A_2, and A_3 are determined by the identity

$$\frac{2\gamma p^2 + 4\gamma p + \beta}{\gamma p^3 + 2\gamma p^2 - (\alpha - \beta)p - \alpha} = \frac{A_1}{p - p_1} + \frac{A_2}{p - p_2} + \frac{A_3}{p - p_3} \tag{15}$$

The integration constant C, appearing in equation (13), is determined by equation (9) together with the initial values N_o and p_o at time zero.

It is seen that the values of p_i and A_i ($i = 1, 2, 3$) are functions of the two ratios

$$a = \alpha/\gamma \quad , \qquad b = \beta/\gamma$$

only. Further, if $b \lesssim 1$, a condition which is always fulfilled according to the data published by Bates et al. [3] and Collins [6], all the zeros p_i are real: one is positive and two are negative. Equation (9) can be written

$$\dot{p} = -\gamma N(p - p_1)(p - p_2)(p - p_3) \tag{16}$$

where p_1 is the positive solution. The factors $(p - p_2)$ and $(p - p_3)$ are then positive, since the density ratio p, as well as γ and N, are positive quantities. Hence,

$$\dot{p} > 0 \text{ if } p < p_1, \quad \text{and} \quad \dot{p} < 0 \text{ if } p > p_1,$$

and during the whole afterglow, p tends to approach the positive solution p_1. Computed values of p_i and A_i are given in tables 1-3 for different values of a and b. The following may be noted:

for $a \gg 1$, $b \leq 1$ $\quad p_1 \rightarrow \sqrt{a}$ (17)

for $a \ll 1$, $b \lesssim a$ $\quad p_1 \rightarrow \sqrt{a/2}$ (18)

for $a \ll 1$, $b \gg a$ $\quad p_1 \rightarrow a/b = \alpha/\beta$ (19)

and, in addition $\quad A_1 \gtrsim 1$ (20)

$$A_1 + A_2 + A_3 = 2 \tag{21}$$

$$p_1 + p_2 + p_3 = -2 \tag{22}$$

Table 1. b = 0

a	p_1	p_2	p_3	A_1	A_2	A_3
0.0191	0.1	-0.10	- 2.00	1.022	0.973	0.005
0.0733	0.2	-0.18	- 2.02	1.039	0.942	0.018
0.417	0.5	-0.40	- 2.10	1.071	0.830	0.098
1.50	1.0	-0.63	- 2.37	1.091	0.612	0.297
5.33	2.0	-0.85	- 3.15	1.091	0.297	0.612
29.2	5.0	-0.97	- 6.03	1.063	0.066	0.871
109	10.0	-0.99	-11.01	1.039	0.018	0.942

Table 2. b = 0.5

a	p_1	p_2	p_3	A_1	A_2	A_3
0.0645	0.1	-0.37	- 1.73	1.063	1.117	- 0.181
0.157	0.2	-0.45	- 1.75	1.092	1.051	- 0.143
0.583	0.5	-0.62	- 1.88	1.125	0.860	0.015
1.75	1.0	-0.79	- 2.21	1.130	0.558	0.312
5.67	2.0	-0.92	- 3.08	1.112	0.236	0.652
29.6	5.0	-0.98	- 6.02	1.070	0.050	0.881
110	10.0	-1.00	-11.00	1.041	0.014	0.945

Table 3. b = 1.0

a	p_1	p_2	p_3	A_1	A_2	A_3
0.110	0.1	-1.00	- 1.10	1.076	9.091	-8.167
0.240	0.2	-1.00	- 1.20	1.119	4.167	-3.286
0.750	0.5	-1.00	- 1.50	1.167	1.333	-0.500
2.00	1.0	-1.00	- 2.00	1.167	0.500	0.333
6.00	2.0	-1.00	- 3.00	1.133	0.167	0.700
30.0	5.0	-1.00	- 6.00	1.076	0.033	0.891
110	10.0	-1.00	-11.00	1.043	0.009	0.948

When the density ratio p is closely equal to p_1, we have what is generally called a "transient equilibrium" between the concentrations of free electrons and metastables. The rate of plasma decay under this condition will be discussed below. First, however, we shall calculate the time of transition from the initial value p_o to the asymptotic value p_1. It will be shown that this transition time is much shorter then the characteristic decay time for the electron density.

4. THE TRANSITION TIME

Equation (13) for p can be solved exactly, provided that the exponents A_i are integers. One such case is in the limit of large a, for all values of $b \leq 1$, where

$$A_1 = 1, \qquad A_2 = 0, \qquad A_3 = 1$$

Comparing equations (13) and (16) one finds

$$C = - \frac{\gamma N_o (p_o - p_1)}{|p_o - p_1|^{A_1} (p_o - p_2)^{A_2 - 1} (p_o - p_3)^{A_3 - 1}} \tag{23}$$

where N_o and p_o are taken at the time t_o. Hence,

$$C < 0 \quad \text{if} \quad p_o > p_1$$

and $$C > 0 \quad \text{if} \quad p_o < p_1$$

For large a, equation (13) may then be written

$$\dot{p} = - |C| (p - p_1)(p - p_3) \tag{24}$$

with the solution

$$\frac{p - p_1}{p - p_3} = \frac{p_o - p_1}{p_o - p_3} e^{-(t - t_o)/\tau} \tag{25}$$

and where the characteristic time τ is

$$\tau = \frac{1}{(p_1 - p_3)|C|} \tag{26}$$

Inserting asymptotic values for p_i, this expression becomes

$$\tau = \frac{1}{2\gamma N_o (p_o + 1)\sqrt{a}} = \frac{1}{2\sqrt{\alpha\gamma}(M_o + N_o)} \tag{27}$$

Also when a and b are both small, the exponents A_i are integers:

$$A_1 = 1, \qquad A_2 = 1, \qquad A_3 = 0$$

The solution for p is then again given by the expression (25), with p_3 replaced by p_2, and with

$$\tau = \frac{1}{(p_1 - p_2)\,|C|} = \frac{1}{\gamma N_o (p_o + 2)\,\sqrt{2}a} = \frac{1}{\sqrt{2\alpha\gamma}(M_o + 2N_o)} \tag{28}$$

It is seen that the characteristic transition times τ for these two considered cases, given by the expressions (27) and (28), respectively, differ by at most a factor $\sqrt{2}$: this maximum difference occurs for $p_o = 0$.

In the general case, when A_i are not integers, it is convenient to define a characteristic transition time for the quasi-exponential approach in time of p to the asymptotic value p_1:

$$\tau = - \frac{p - p_1}{\dot{p}} \tag{29}$$

We want to evaluate this transition time for the situation where the density ratio p is only "slightly" deviating from the "transient equilibrium" value p_1, which could occur for instance when the rate coefficients α, β, and γ vary slowly in time. Inserting the expressions (13) and (23), and approximating $A_1 = 1$, $p = p_1$, we obtain

$$\tau = \frac{1}{N_o \sqrt{\alpha\gamma}}\, f(a,\ b,\ p_o) \tag{30}$$

where the function $f(a, b, p_o)$ is given by

$$f(a,\ b,\ p_o) = \frac{\sqrt{a}}{(p_o - p_2)^{1-A_2}\,(p_o - p_3)^{1-A_3}\,(p_1 - p_2)^{A_2}\,(p_1 - p_3)^{A_3}} \tag{31}$$

It is easily verified, by inspection of tables 1-3, that the expressions (30) and (31) develop into the expressions (27) and (28) for the two respective special cases considered above. For fixed values a and b, τ has its maximum value for $p_o = 0$: this value then represents an upper bound for the transition time τ at those particular values a, b, γ, and N_o. The function $f(a, b, p_o = 0)$ is shown graphically in Fig. 1: its variation is quite small, and

$$f(a, b, 0) \lesssim 1/2$$

Hence

$$\tau \leq \frac{1}{2N_o\sqrt{\alpha\gamma}} \tag{32}$$

From equation (16) τ may be expressed in terms of the instantaneous electron density N, if again p is nearly equal to p_1:

$$\tau = \frac{1}{\gamma N(p_1 - p_2)(p_1 - p_3)} \tag{33}$$

Rewriting this expression as

$$\tau = \frac{1}{\gamma N(a + \sqrt{a})}\, g(a, b) \tag{34}$$

it is found that the function

$$g(a, b) = \frac{a + \sqrt{a}}{(p_1 - p_2)(p_1 - p_3)} \tag{35}$$

is very similar to the function $f(a, b, p_o = 0)$, in particular it varies slowly with a and b. Common features of the two functions are

$$f,\ g \longrightarrow 1/2 \qquad \text{for } a \longrightarrow \infty \tag{36}$$

$$f,\ g \lesssim 1/2 \tag{37}$$

$$f,\ g \longrightarrow \sqrt{2}/4 \qquad \text{for } a \longrightarrow 0,\ b = 0 \tag{38}$$

$$f,\ g \propto \sqrt{a} \qquad \text{for small a, } b = 1 \tag{39}$$

From the upper bound of g it follows that

$$\tau \leq \frac{1}{2\gamma N(a + \sqrt{a})} = \frac{1}{2N(\alpha + \sqrt{\alpha\gamma})} \tag{40}$$

In a helium afterglow experiment, the reaction rate coefficients α, β, and γ cannot be expected to be time-independent constants. However, their time variation is quite slow, and in any case not faster than the variation in electron density, which might be expressed as a characteristic time for quasi-exponential decay (during a short time interval):

$$\tau_N \gtrsim -\frac{N}{\dot{N}} = \frac{1}{\alpha_{eff} N} \geq \frac{1}{\alpha N} = \frac{1}{a\gamma N}$$

it is seen, by comparison with the expression (40), that

$$\tau \ll \tau_N$$

In general, between five and ten characteristic times for approaching "transient equilibrium" between the densities of metastables and free electrons have elapsed before the electron density has decayed to 1/e of its initial value N_o. This also means that, even in an afterglow, where the recombination coefficient varies in time, the density ratio p between metastables and free electrons relaxes to a value close to the transient equilibrium value p_1, which is a function of the rate coefficients α, β, and γ only. The rate of plasma decay under such a transient equilibrium will next be discussed.

5. THE EFFECTIVE RECOMBINATION COEFFICIENT

The free electron density decays at a rate given by the equations (8) or (10). Under "transient equilibrium" the positive solution p_1 of equation (14) is to be inserted for the density ratio p in the expression (10). The resulting effective recombination coefficients α_{eff}, as de-

fined by the expression (11), are presented in Fig. 2 as the ratio α_{eff}/α versus $a = \alpha/\gamma$, and with $b = \beta/\gamma$ as parameter.

It is especially noted that, for small a, $b \neq 0$, the ratio α_{eff}/α tends to unity, exactly like the result obtained under the quasi-equilibrium approximation for the metastable density: for the expression (19) is valid in both cases. For larger values of the ratio $a = \alpha/\gamma$, however, it is easily seen from the rate equation (7) that, since the time derivative $\dot{M}$ in reality is negative and different from zero, the metastable density M must be larger than the quasi-equilibrium value obtained from the approximation $\dot{M} = 0$. Thus, for large a the ratio α_{eff}/α decreases like $1/\sqrt{a}$, while in the quasi-equilibrium approximation it approaches the value 1/2.

Inserting experimental values of β and γ for helium, reported by Bates et al. [3] and Collins [6]:

$$\beta = 6.4 \cdot 10^{-16}\ m^3/sec, \qquad \gamma = 1.85 \cdot 10^{-15}\ m^3/sec$$

it is seen that the ratio α_{eff}/α is reduced to about one half even for recombination coefficients α as small as $10^{-15}\ m^3/sec$. This implies that ionizing collisions between metastable atoms or molecules are of importance in practically all laboratory helium afterglow plasmas, a fact which must be taken into account in any experimental determination of the recombination coefficient for helium ions, based upon afterglow measurements of the decay of electron density.

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CAPTIONS OF FIGURES

Fig. 1 A graph of the function $f(a, b, p_o = 0)$ versus a with b as parameter.

Fig. 2 A graph of the recombination coefficient ratio α_{eff}/α versus a with b as parameter.

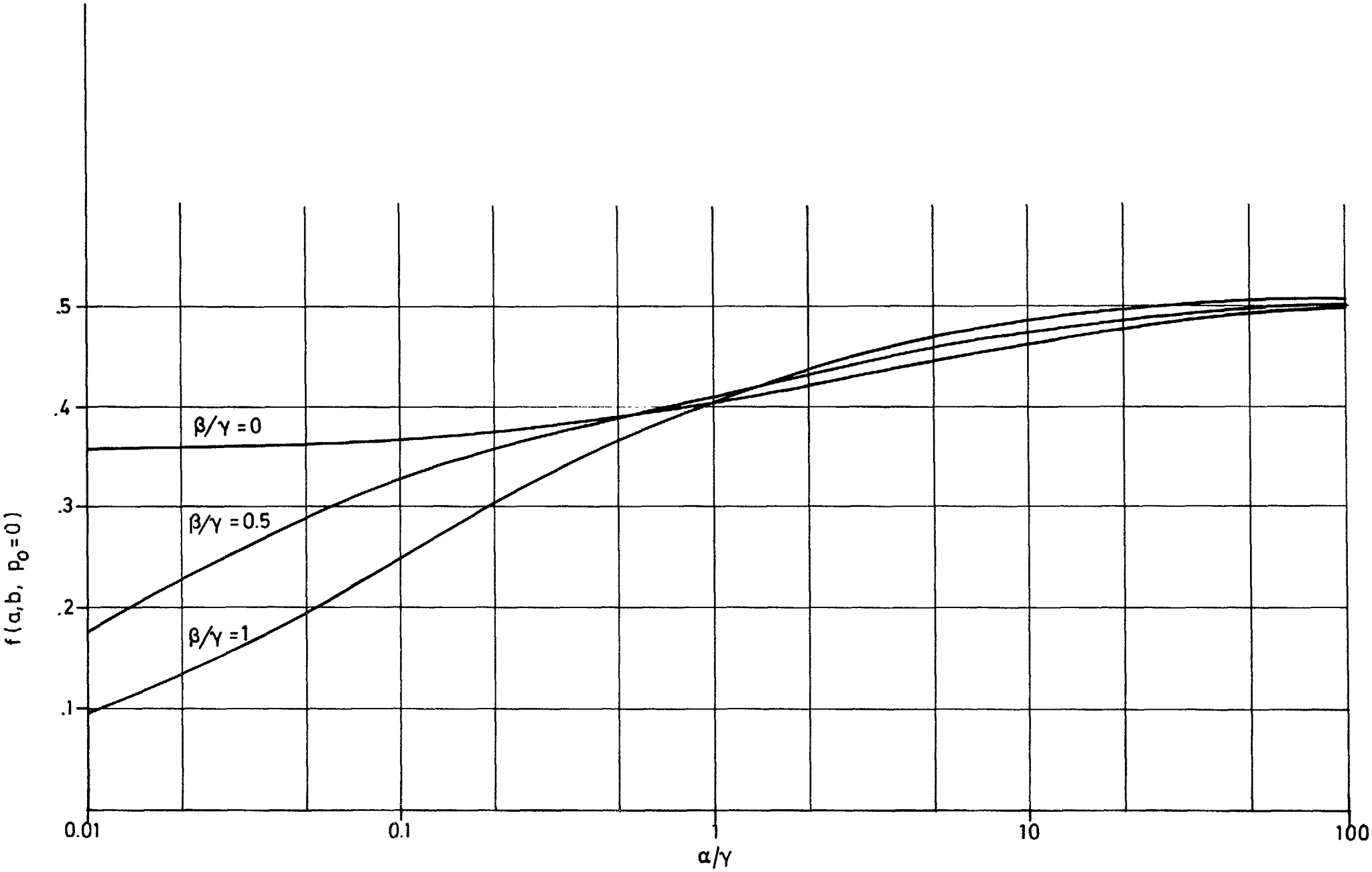

FIGURE 1

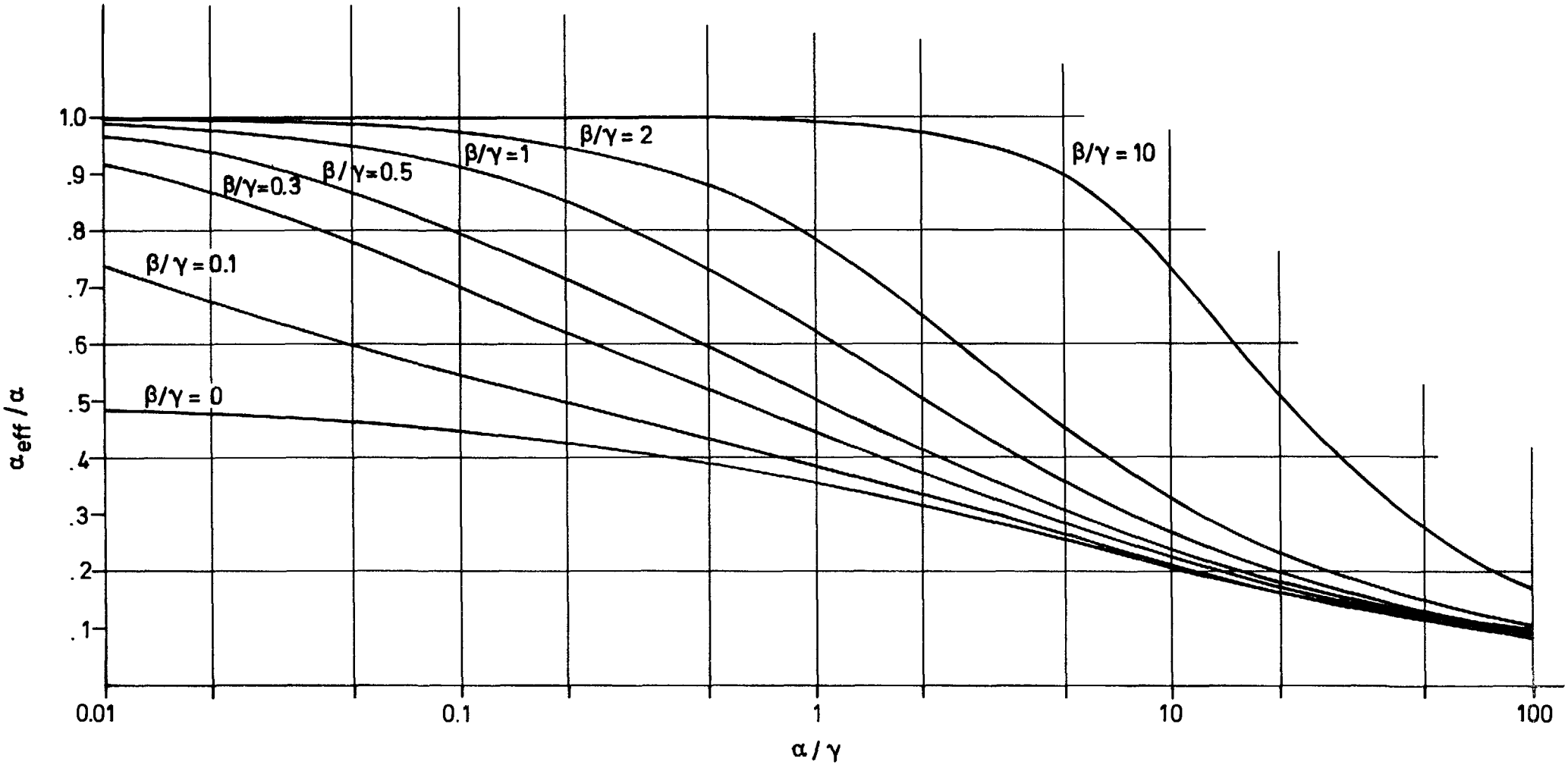
1.0
.9
.8
.7
.6
.5
.4
.3
.2
.1
α_{eff}/α
β/γ = 2
β/γ = 1
β/γ=0.5
β/γ=0.3
β/γ = 0.1
β/γ = 0
β/γ = 10
0.01
0.1
1
10
100
α/γ

FIGURE 2

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