NEGATIVE ABSORPTION COEFFICIENT PRODUCED BY DISCHARGES IN A GAS MIXTURE

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The probabilities for various processes are considered in an analysis of an electrical discharge in a mixture of two gases in which a negative absorption coefficient is obtained by selective excitation of atoms to a higher level and selective removal of atoms from a lower level.

1. Basov and Krokhin[1] have proposed a diagram that facilitates analysis of the necessary conditions for obtaining a negative absorption coefficient in an electrical discharge in a mixture of two gases.*

The assumptions used by these authors, however, correspond neither to actual conditions nor to the relations between atomic constants for vapors and gases used by Butaeva and Fabrikant[2] and by Ablekov, Pesin and Fabelinskii.[3] In [2] collisions between atoms and molecules of two components of a gas mixture were used to selectively remove atoms from a lower level; however, atoms were not selectively excited to an upper level.[1] This technique had been proposed in 1939 and is the original prototype of the method used in ammonia masers. This case is the opposite of that considered by Basov and Krokhin.

Entirely different results are obtained if calculations similar to those in [1] are used for the conditions pertaining to [2]. In particular, inequality (5) of [1] becomes completely different and the corresponding diagram contains only two regions, I and II; in contrast with [1], region IV, in which a negative absorption coefficient is impossible, no longer appears. It should also be noted that in [2] the discharge conditions were such that the Maxwellian electron energy did not appear explicitly, so that the relation between Θ_{i0} and Θ_{0i} given in [1] does not hold.

In [1] account is taken of collisions of the second kind with electrons in which atoms are transferred from level ϵ_i to level ϵ_0 while collisions of the second kind characterized by $\epsilon_i \rightarrow \epsilon_k$ are neglected. The principle of detailed balancing shows that the probability of collisions of the second kind increases as the distances between levels decrease.[5] Consequently the probability of $\epsilon_i \rightarrow \epsilon_k$ collisions of the second kind is greater than the probability of $\epsilon_i \rightarrow \epsilon_0$ transitions. Also, the quantity $1/\tau_{ik}$ cannot be neglected if $1/\tau_{i0}$ is retained. For example, in the case of mercury and zinc $1/\tau_{i0}$ is approximately 0 while $1/\tau_{ik} \approx 10^7$ sec^{-1}. The same situation holds for the inert gases.[6] It is well known that spontaneous transitions characterized by $\epsilon_i \rightarrow \epsilon_k$ in the optical frequency region generally have a strong inhibiting effect on the establishment of the required relation between the atomic populations in levels ϵ_i and ϵ_k.

*The term "negative absorption coefficient" corresponds more closely to the terminology used by Einstein than the presently popular term "negative temperature."

2. We now derive the conditions that must be satisfied to obtain a negative absorption coefficient when the actual relations between the appropriate probabilities are taken into account. We first form the equations that describe selective removal of atoms from the lower level. Aside from a few changes our notation is essentially that used earlier in the theory of the gas discharge.[7] Thus, we have

$$dn_i^a/dt = -n_i^a(A_{ik}+\beta_{ik}) + n_0^a\alpha_{0i} + n_k^a\alpha_{ki}, \tag{1}$$

$$dn_k^a/dt = -n_k^a(A_{k0}+\beta_{k0}+B_{ab}) + n_0^a(\alpha_{0k}+B_{ba}) + n_i^a(A_{ik}+\beta_{ik}), \tag{2}$$

where n is the density of atoms or molecules; A_{ik} and A_{k0} are the probabilities for spontaneous transitions; $B_{ab} = n_0^b\sigma_{ab}v$, B_{ba} is the probability for collisions with impurity atoms or molecules (the impurity is the added component of the mixture); α_{0i}, α_{0k}, α_{ki} are the probabilities for electron collisions of the first kind; β_{i0}, β_{k0}, β_{ik} are the probabilities for electronic collisions of the second kind.

In writing Eqs. (1) and (2) we assume that

$$A_{i0} \ll A_{ik}, \ \alpha \ll \beta, \ \beta_{i0} \ll \beta_{ik}.$$

The lifetimes are written in the form

$$\tau_i = 1/(A_{ik} + \beta_{ik}), \quad \tau_k = 1/(A_{k0} + \beta_{k0} + B_{ab}). \quad (3)$$

Using Eqs. (1) and (2) we obtain the condition for producing a negative absorption coefficient in the steady state:

$$\frac{\alpha_{0i}(A_{k0} + \beta_{k0} + B_{ab}) + \alpha_{ki}(\alpha_{0k} + B_{ba})}{(\alpha_{0k} + \alpha_{0i} + B_{ab} n_k^b / n_0^b)(A_{ik} + \beta_{ik})} > 1. \quad (4)$$

Using Eq. (3) we can show that (4) is an extended version of inequality (10) of [2].

Using the approximations

$$\beta_{ik} \approx b_{ik}\beta_{i0}, \quad \beta_{k0} \approx b_{k0}\beta_{i0}, \quad (5)$$

where b_{ik} and b_{k0} are essentially constants appreciably greater than unity, we obtain from Eq. (4)

$$\frac{B_{ab}}{\beta_{k0}} \gtrless \frac{b_{ik}[\exp\{(\varepsilon_i - \varepsilon_k)/kT_e\} + 1/b_{k0}]\eta_{ik} - \eta_{k0} - b_{ik}}{1 - b_{ik}(n_k^b/n_0^b)[\exp\{\varepsilon_i/kT_e\}\eta_{ik} - \exp\{\varepsilon_k/kT_e\}]} \quad (6)$$

(we have used the notation $\eta_{ik} \equiv 1 + A_{ik}/\beta_{ik}$). The > sign applies when the denominator is greater than zero while the < sign applies when the denominator is less than zero.

The equations for the lines on the A_{k0}/β_{k0}; A_{ik}/β_{ik} diagram are of the form

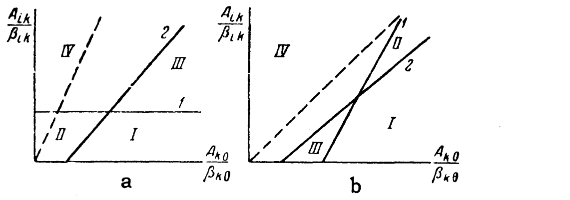
$$\frac{A_{ik}}{\beta_{ik}} = \frac{\exp\{-\varepsilon_i/kT_e\}}{b_{ik}} \frac{n_0^b}{n_k^b} - 1 - \exp\{-(\varepsilon_i - \varepsilon_k)/kT_e\}, \quad (7)$$

$$\frac{A_{ik}}{\beta_{ik}} = \frac{A_{k0}/\beta_{k0} + 1 - b_{ik}(1/b_{k0} - 1 + \exp\{(\varepsilon_i - \varepsilon_k)/kT_e\})}{b_{ik}(\exp\{(\varepsilon_i - \varepsilon_k)/kT_e\} + 1/b_{k0})}. \quad (8)$$

Equation (7) is physically meaningful only when

$$n_k^b/n_0^b < \exp\{-\varepsilon_i/kT_e\}/b_{ik}. \quad (9)$$

The inequality (9) is certainly not always satisfied in a gas discharge. In Eq. (8) the free term



a – selective removal of atoms from the lower level; 1 – line (7), 2 – line (8); b – selective excitation of atoms to the upper level; 1 – line (14), 2 – line (15). Region I – impurity desirable, II – impurity necessary, III – impurity harmful, IV – effect impossible.

is always negative, in contrast with the case considered in[1]. If the condition in (9) is satisfied the diagram will contain four regions (cf. Fig. a) characterized by the following features: I) an impurity is not necessary, but is desirable; II) an impurity is necessary; III) an impurity is harmful; IV) it is impossible to obtain a negative absorption coefficient. Regions I and II vanish if the condition in (9) is not satisfied.

We now consider selective excitation of atoms to a level ϵ_i. In this case the starting equations are

$$dn_i^a/dt = -n_i^a(A_{ik} + \beta_{ik} + B_{ab}) + n_0^a(\alpha_{0i} + B_{ba}) + n_k^a\alpha_{ki}, \quad (10)$$

$$dn_k^a/dt = -n_k^a(A_{k0} + \beta_{k0}) + n_0^a\alpha_{0k} + n_i^a(A_{ik} + \beta_{ik}). \quad (11)$$

The condition for a negative absorption coefficient becomes

$$\frac{(\alpha_{0i} + B_{ba})(A_{k0} + \beta_{k0}) + \alpha_{0k}\alpha_{ki}}{(A_{ik} + \beta_{ik} + B_{ab})\alpha_{0k} + (A_{ik} + \beta_{ik})(\alpha_{0i} + B_{ba})} > 1. \quad (12)$$

The inequality (12) also represents an extended form of (10) of [2]. Consequently

$$\frac{B_{ba}}{\beta_{i0}} \gtrless \frac{(b_{ik}/b_{k0})[\eta_{ik}(1 + b_{h0}\exp\{(\varepsilon_i - \varepsilon_k)/kT_e\}) - (b_{k0}/b_{ik})\eta_{k0} - b_{ik}\exp\{(\varepsilon_i - \varepsilon_k)/kT_e\}]}{\exp\{\varepsilon_i/kT_e\}[\eta_{h0} - (b_{ik}/b_{k0})[\eta_{ik} - (n_0^b/n_i^b)\exp\{-\varepsilon_k/kT_e\}]]}. \quad (13)$$

The equations for the lines on the A_{k0}/β_{k0}; A_{ik}/β_{ik} diagram are of the form

$$\frac{A_{ik}}{\beta_{ik}} = \frac{b_{k0}}{b_{ik}}\frac{A_{k0}}{\beta_{k0}} - \frac{b_{k0}}{b_{ik}}\left(\frac{n_0^b}{n_i^b}\exp\left\{-\frac{\varepsilon_k}{kT_e}\right\} - 1\right) - 1, \quad (14)$$

$$\frac{A_{ik}}{\beta_{ik}} = \frac{A_{k0}/\beta_{k0} - b_{ik}\exp\{(\varepsilon_i - \varepsilon_k)/kT_e\}(1 - 1/b_{k0}) - b_{ik}/b_{k0} + 1}{b_{ik}\exp\{(\varepsilon_i - \varepsilon_k)/kT_e\} + b_{ik}/b_{k0}}. \quad (15)$$

The free terms in Eqs. (14) and (15) are always negative.

The lines intersect when

$$n_i^b/n_0^b < \exp\{-\varepsilon_i/kT_e\}/b_{ik}. \quad (16)$$

In this case the diagram becomes that shown in Fig. b, where the shapes of the various regions are different from those in the diagram in [1]. If the excitation of impurity atoms becomes strong enough, the condition in (16) is violated and region III disappears.

Because of (5), for a given host material transfers are possible only in regions of the diagrams close to the dashed line. The slope of these lines is determined by the quantity $b_{k0}A_{ik}/b_{ik}A_{k0}$.

The value of diagrams of this kind is limited by the fact that the lines which divide the diagrams into various regions are displaced sharply for small changes in the electron temperature. In particular, shifts of this kind occur when the composition of the medium is changed in order to vary the quantity B_{ab}.

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