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Fig. 1

Figure 1: Fig. 1

Abstract

Full Text

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PHYSICS

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On the Role of Molecular-Ionic Formations in the Excitation of Spectral Lines of Atoms

In a recently published note by I. P. Bogdanova and I. Geits (^{1}), it was shown that, on the optical excitation functions of certain helium lines, an additional maximum arises near the threshold if hydrogen, krypton, or mercury vapor is added to the helium. When neon is added, the appearance of an additional maximum is not observed.

At present the following additional experiment has been carried out: the electric field accelerating the electrons in the beam was applied in the form of short pulses of duration $\sim 10^{-7}$ sec, separated by intervals of $2 \cdot 10^{-5}$ sec. The intensity of the excited helium lines was measured with a photomultiplier operating in the photon-counting mode, by the method of correlated coincidences. The measurement was made only during the application of the pulse accelerating the electrons. In Fig. 1 the solid curve gives the optical excitation function obtained in this way for the He I line λ 4713 Å with xenon added ($p_{\mathrm{Xe}} = 3 \cdot 10^{-3}$ mm Hg; $p_{\mathrm{He}} = 2 \cdot 10^{-2}$ mm). The dashed line depicts the optical excitation function measured with a continuously acting accelerating field. As can be seen, during the short excitation time ($\sim 10^{-7}$ sec), the additional maximum does not have time to form. This indicates that the appearance of the additional maximum is connected with a process developing in time more slowly than the process of excitation of an atom by electron impact followed by spontaneous radiation.

Fig. 1

A process leading to this kind of "delayed" excitation of atomic levels may be a process connected with deionization of an atomic ion, or with the appearance and subsequent decay of a molecular or molecular-ionic formation.

Yu. M. Alenskovskii and V. L. Granovskii (2), who investigated the decay of helium plasma, considered the process of recombination of atomic ions in triple collisions with electrons. However, such a process cannot explain the occurrence of the secondary maximum, since this maximum arises at energies lower than the ionization energy. Also little probable is a triple-collision process leading to the formation of the molecule He_2 and its subsequent dissociation.

As a process arising in double collisions and leading to the formation of the additional maximum, one may indicate the following process, considered by Molnar and Hornbeck (3):

$$\mathrm{He} + e \to \mathrm{He}^{*} + e; \tag{1}$$

$$\mathrm{He}^{*} + \mathrm{He} \to \mathrm{He}_2^{+} + e'; \tag{2}$$

$$\mathrm{He}_2^{+} + e \to \mathrm{He}^{*} + \mathrm{He}. \tag{3}$$

The sign $*$ denotes the excited state of an atom. Thus, it follows that a helium atom excited by an electron (process (1)), colliding during its lifetime with a normal helium atom, forms a molecular ion (process (2)). The collision of the latter with a **slow** electron leads to dissociative recombination with the formation of two neutral atoms, one of which is excited (process (3)). Dissociative recombination leads to **selective** excitation of atoms (mainly S- and D-levels). This explains why an additional maximum is observed on the optical excitation functions not for all helium lines. The role of impurities ($H_2, \mathrm{Kr}, \mathrm{Hg}$) is secondary: it leads to the appearance in the beam of slow electrons, which was confirmed by direct measurements.

The role of dissociative recombination of He_2^{+} ions apparently also explains the features in the change of the helium spectrum upon transition from a "weak" to a "strong" high-frequency discharge. In the work of O. P. Bochkova and L. P. Razumovskaya (4) it was shown that, upon transition from a "weak" to a "strong" discharge, there occurs a stepwise change in the concentration of free electrons in the discharge plasma without a noticeable change in the electron temperature. At the same time, the intensity of helium lines increases, and not equally for lines with different upper levels. Calculations performed under the assumption of purely electronic excitation of the levels and their decay due to spontaneous transitions show that the population of high atomic levels both in the "weak" and in the "strong" discharge is smaller than that which should have corresponded to the measured electron concentrations and electron temperature. Smaller-than-calculated concentrations of excited neon levels were also observed in the positive column of a dc discharge by a number of authors. Yu. M. Kagan, R. I. Lyagushchenko, and A. D. Khakhaev (5) explain this discrepancy by stepwise ionization. However, under the conditions in which the transition from a "weak" to a "strong" discharge occurs, stepwise ionization cannot play an essential role. Another cause must be sought; such a cause may be the formation of molecular He_2^+ ions in accordance with reaction (2). In the "strong" discharge this process proceeds more intensively, and at the same time dissociative recombination begins to play a role, leading to selective population of some helium levels.

The indicated scheme makes it possible to estimate the effective cross sections of processes (2) and (3). Let us define the cross section Q_2 of process (2) by the relation

$$\Delta N_k' = N_0 N_k \bar{v} Q_2, \tag{4}$$

where $\Delta N_k'$ is the number of destruction events per unit volume per unit time of the k-th excited level of the helium atom; N_0 and N_k are the concentrations of normal and excited atoms; $\bar{v}$ is their mean relative velocity. Assuming excitation to be electronic, we obtain the stationarity condition:

$$\Delta N_{\text{el}} = \Delta N_k' + N_k \sum_i A_{ki}, \tag{5}$$

where ΔN_{el} is the number of excitation events of the k-th level due to collisions with electrons; A_{ki} are the transition probabilities. The concentration of excited atoms entering into expression (5) corresponds to the actually observed one; therefore we shall denote it by $N_{k\,\text{obs}}$. The concentration that would occur in the absence of process (2) we shall denote by $N_{k\,\text{calc}}$; it is determined by the equality

$$\Delta N_{\text{el}} = N_{k\,\text{calc}} \sum_i A_{ki}. \tag{5a}$$

From (5) and (5a) we obtain

$$\Delta N_k' = \Delta N_{\text{el}} \left(1 - \frac{N_{k\,\text{obs}}}{N_{k\,\text{calc}}}\right). \tag{6}$$

For high levels, according to the experimental data, $N_{k\,\text{obs}}/N_{k\,\text{calc}} \simeq 0.05$; hence, from (6), it follows that $\Delta N_k' \simeq \Delta N_{\text{el}}$. The number of electron excitations ΔN_{el} is calculated from the known effective cross sections for excitation of helium-atom levels by collisions with electrons (6). After this, from relation (4) (using N_0 and N_k corresponding to the conditions of our experiment) we obtain: $Q_2 \simeq 2 \cdot 10^{-15}$ cm^2.

The effective cross section Q_3 of process (3) can be estimated on the basis of the relation

$$\Delta N'' = N_+ n'_e \overline{v}_e Q_3, \tag{7}$$

where $\Delta N''$ is the number of acts of dissociative recombination; N_+ is the concentration of molecular ions; n'_e is the concentration of slow electrons (energy $\ll 2$ eV); $\overline{v}_e$ is their mean velocity. To estimate the concentration N_+, we assume that in the "weak" discharge it is negligibly small. Then for the "strong" discharge, owing to the quasineutrality of the plasma, one may take $N_+ = \Delta n_e$, where Δn_e is the increase in the concentration of free electrons upon transition from the "weak" to the "strong" discharge. Further, under stationarity of the process,

$$\Delta N_{\text{el}} - \Delta N' + \Delta N'' = N_k \sum_i A_{ki}. \tag{8}$$

From (7) and (8) we find for Q_3 a value of the order of 10^{-13} cm^2. The cited values of the cross sections Q_2 and Q_3 should be regarded as their upper limit, since they were calculated with neglect of a number of other processes, besides processes (2) and (3), which may also play a role in the population and destruction of helium levels.

In conclusion we note that A. M. Shukhtin, V. S. Egorov, and Yu. G. Kozlov (7), on the basis of their observations of the excitation of neon atoms during the decay of a plasma containing a neon–helium mixture, also arrive at the conclusion that molecular-ion formations and their decay play a major role in the process of atomic excitation. Apparently, selective population of excited atomic levels in dissociative recombination may play a substantial role in creating population inversion. It has recently been shown (8) that analysis of the conditions for the appearance of population inversion in a neon–helium laser leads to the conclusion that this process is not provided by second-kind collisions between excited helium atoms and normal neon atoms. It is possible that here as well (as in lasers operating on pure inert gases) a process analogous to the one analyzed occurs.

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Note: Figure translations are in progress. See original paper for figures.

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