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ELECTRON-ION RECOMBINATION IN THE TRACK OF AN IONIZING PARTICLE AND THE SCINTILLATION MECHANISM OF NOBLE GASES

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The role of electron-ion recombination in the scintillation of noble gases is established experimentally. A scintillation mechanism based on the formation, emission, and de-excitation of noble-gas molecules in the ionizing-particle track is considered.

1. INTRODUCTION

It is well known that the effectiveness of scintillations of a noble gas under the influence of charged particles can decrease in a weak electric field (gaseous argon[1,2], liquid helium[3], gaseous and liquid xenon[4]).

It is assumed in the cited papers that the decrease of the yield of the scintillation light is due to the decrease of the number of acts of recombination of the electrons with the ions, owing to the departure of some of the electrons from the track of the ionizing particle under the influence of the electric field. It is thus assumed that electron-ion recombination on the track of the ionizing particles is accompanied by emission of light, and consequently it plays an important role in the mechanism of scintillations of noble gases. The hypothesis of electron recombination was also used in other papers to explain the regularities of the glow of gas scintillators (see, for example,[5,6]).

At the same time, the presence of electron-ion recombination in the columns of ionized gas on the particle tracks was confirmed only by the splendid agreement between the experimental results of studies of the yield of electrons from columns in ionization chambers with the Jaffe recombination theory[7]; at the same time, many experiments were performed (for example[8], and earlier investigations reported in the review[9]), which have shown that in gas scintillators placed in an electric field, the yield of the light does not decrease, i.e., there was apparently no recombination. It should also be noted that the experiments described in[1-4], strictly speaking, do not prove the presence of luminescence connected with electron-ion recombination, and indicate only the appreciable role of the electrons in the scintillation process.

In[10] it is proposed that an appreciable contribution to the scintillation may be made by decelerating electrons, produced on the track of the particle, as a result of the bremsstrahlung which appears when the electrons are scattered by neutral atoms[11]. Upon being drawn from the track of the ionizing particle, the electrons lose their ability to draw energy via collisions of the second kind with the excited atoms (molecules) of the gas, and therefore the yield of the scintillation light due to the bremsstrahlung of the electrons should decrease. However, quantitative estimates show[10] that the yield of the bremsstrahlung light should be less than 10% of the observed yield of the scintillation light.

With increasing electric field, the electrons are accelerated and the yield of the light due to the bremsstrahlung of the electrons drifting in the electric field should increase. When the electrons reach the excitation energy of the gas atoms, the electroluminescence may be due to two causes—bremsstrahlung of the electrons and emission of the excited atoms (molecules). The increase of the scintillator glow in the electric field can be readily observed experimentally[12]. We note that electroluminescence can greatly weaken or completely mask the decrease of the yield of the gas scintillator in the electric field.

Thus, it is important to establish the following: 1) the role of electron-ion recombination in processes of atom collisions on the track of an ionizing particle; 2) the contribution of light from electron-ion recombination to the amplitude of noble-gas scintillations.

2. EXPERIMENTAL SETUP

The experimental setup for observing the influence of the electric field on the scintillations of noble gases was essentially an ordinary ionization chamber with flat or cylindrical coaxial electrodes. The scintillation light emerged from the chamber through a window, whose internal surface was coated with a thin semi-transparent layer of quaterphenyl ($\sim 100\ \mu g/cm^2$) to transform the ultraviolet radiation into visible light. The light was registered with a photomultiplier. The ionization pulse and the pulse from the photomultiplier, operating in the regime in which all the light was collected, could be fed simultaneously to an oscilloscope or to a pulse-height analyzer. The chamber was kept in a thermostat. Specially purified noble gases were used. The gas was usually circulated continuously through the chamber and through calcium heated to 500—600°C, thus ensuring high purity of the gas in the chamber.

The scintillations were initiated by α-particles of energy 5.15 MeV or by a pulsed x-ray beam of 300 nsec duration at an average photon energy 60 keV.

3. EXPERIMENTAL RESULTS

A. Two Scintillation Components

Figure 1 shows the dependence of the amplitudes of the scintillations and of the ionization pulse in xenon under the influence of α-particles on the electric-field intensity. It is seen that the decrease of the amplitude of the light pulse in the field reaches ~30%.

If the decrease of the scintillation amplitude is connected with the drawing of the electrons from the α-particle track, then the decrease of the scintillation amplitude and the growth of the ionization-pulse amplitude should be linearly related. The indicated proportionality is illustrated by Fig. 2. Figure 3 shows the dependence of the reciprocal amplitude of the ionization pulse on the reciprocal field intensity. As follows from the Jaffe theory[7], this dependence should be linear if the change of the ion density on the track with time is determined by diffusion, recombination in double collisions, and by drift of the ions in the homogeneous external electric field. The simple form of the ionization curves, in terms of the coordinates used in Fig. 3[13], allows us to extrapolate these curves to infinitely large electric field intensities. The linear connection between the number of drawn-out electrons (amplitude of the ionization pulse) and the decrease of the scintillation light allows us to find the complete decrease of the scintillation light in an infinitely strong field, i.e., that part of the light which is not connected with recombination. The results for α-particles are shown in Fig. 4 (curve 1) at different xenon densities. The figure shows also analogous data for that component of the scintillation pulse amplitude in xenon, which depends on the electric field ("recombination" component)—2, and the summary curve 3.

It is seen from Fig. 4 that the amplitude of scintillations from α-particles can decrease in an electric field by more than a factor of 2 (in Fig. 1 this decrease was masked by electroluminescence). The growth of curves 2 and 3 up to pressures ~30 atm (ρ = 0.2 g/cm^3) is connected with the increased role of the recombination processes in the scintillation. The increase in the role of the recombination processes with increasing gas density is due to the increase of the ionization density on the α-particle track. Therefore, at a fixed gas density, the contribution of the recombination processes to the scintillation should depend on the ionizing ability of the particle.

It is important to note that, in the region where the scintillation amplitude decreases ($\rho > 0.2$ g/cm^3), the decrease of the field-independent component is proportional to the decrease of the "recombination" component, i.e., the mechanism governing the decrease of the scintillation amplitude is apparently the same for both components[1)].

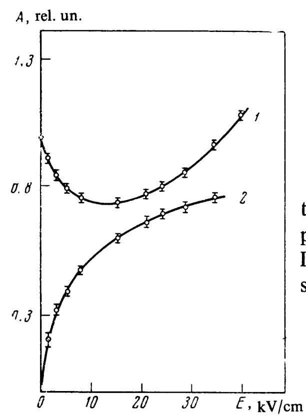

FIG. 1. Amplitude A of scintillations (curve 1) and of the ionization pulse (curve 2) vs. the electric field E. Ionization by α-particles, xenon density 0.66 g/cm^3.

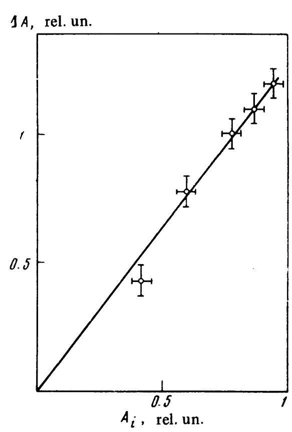

FIG. 2. Decrease of scintillation amplitude ΔA vs. amplitude of ionization pulse A_i. Ionization by α-particles, xenon density 0.66 g/cm^3.

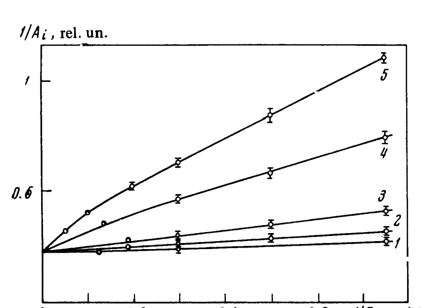

FIG. 3. Dependence of the reciprocal amplitude of the ionization pulse $1/A_i$ on the reciprocal electric field intensity $1/E$ in xenon ionized by α-particles. The curves are obtained at xenon densities: 1–0.125, 2–0.205, 3–0.325, 4–0.495, 5–0.655 g/cm^3.

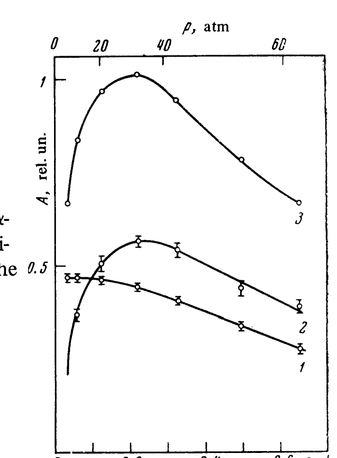

FIG. 4. Dependence of the scintillation amplitude A on the xenon density ρ. Ionization with α-particles: 1—the scintillation amplitude component independent of the field, 2—the field-dependent component of the scintillation amplitude, 3—total scintillation amplitude.

1)The decrease of the amplitude of the scintillations at densities $\rho >$ > 0.20 g/cm^3 cannot be attributed to Rayleigh scattering, since a special experiment has shown that the decrease of the amplitude of the scintillations with increasing density does not depend on the gas temperature.

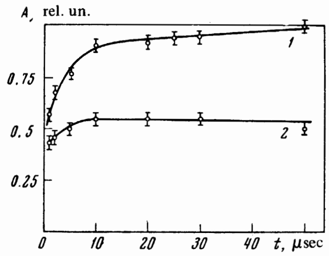

FIG. 5. Growth of the amplitude A of the scintillation pulse with time. Xenon (ρ = 0.3 g/cm^3) ionized with an x-ray pulse. Curve 1—electric field off, curve 2—electric field of 3 kV/cm turned on.

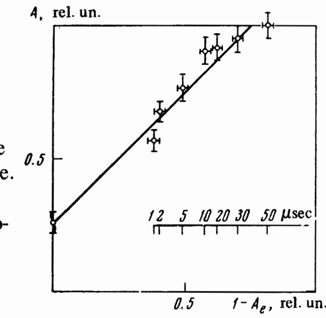

FIG. 7. Dependence of the scintillation amplitude A on the decrease of the electroluminescence amplitude. The points were obtained for different instants of time following the ionization of xenon by x-rays.

Figure 5 shows the growth of the amplitude of scintillations with time when xenon is excited by an x-ray pulse. We see that the scintillations are characterized by large emission times (~10 μsec), so that the origin of the scintillation light cannot be alone attributed to the emission of the excited atoms (molecules, ions) of the gas, produced at the instant of passage of the ionizing particle. When the electric field is turned on, the pulse-amplitude growth time decreases appreciably. This shows that the field-independent scintillation component is emitted much more rapidly.

B. Recombination Glow

Thus, none of the foregoing facts contradict the hypothesis that the electron-ion recombination is accompanied by luminescence on the track of the ionizing particle. To prove that electron-ion recombination produces glow of the gas, it is necessary to show that the number of photons emitted by the gas is proportional to the number of recombination acts. A special experiment was performed for this purpose.

Gas (xenon) was ionized by a pulse of x-radiation, after which a pulsed electric field of large intensity (up to 120 kV/cm) was applied to the electrodes of the chamber. The electrons that escaped recombination prior to the instant of application of the high-voltage pulse, were accelerated in the electric field and the photomultiplier registered a flash of electroluminescence light, proportional to the number of accelerated electrons.

Figure 6 shows the dependence of the electroluminescence light on the delay time between the high-voltage pulse and the instant of passage of the x-radiation. Obviously this curve shows the time dependence of the number of electrons on the particle track. The decrease of the curve is characterized by the rate of the recombination processes on the track. From curves similar to that shown in Fig. 6, we see that the recombination occurs within times ~10 μsec in a broad interval of gas densities (0.1–0.6 g/cm^3).

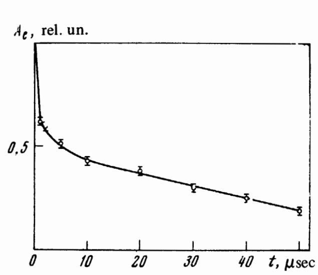

FIG. 6. Decrease of electroluminescence amplitude A_e with time. Xenon (ρ = 0.3 g/cm^3) ionized by an x-ray pulse.

The data shown in Figs. 5 and 6 make it possible to construct the dependence of the scintillation amplitude on the decrease of the amplitude of the electroluminescence at different instants of time after passage of the ionizing radiation (Fig. 7). The electroluminescence amplitude at the zero instant of time is taken to equal unity.

Thus, Fig. 7 shows, in relative units, the connection between the number of photons emitted by the gas at different instants of time and the fraction of the recombining electrons. In the presence of recombination light, the experimental points should lie on a straight line. The intercept of the line on the ordinate axis gives the relative amplitude of the field-independent component of the scintillations. This value was obtained independently by the method described in Sec. 3 A, and is shown in Fig. 7.

Thus, the experimental data confirm that the scintillation amplitude is proportional to the number of recombination acts on the track of the ionizing particle.

C. Spectral Composition of the Scintillations

Since more than 90% of the light of scintillations in a noble gas lies in the region of the vacuum ultraviolet (the scintillation amplitude decreases by 10–15 times if the light is not converted with the aid of quaterphenyl), the spectral measurements entail great difficulty. We determined the characteristic wavelength of the scintillations by means of the photoabsorption of the light by oxygen, which was specially introduced in the volume of the chamber. When a molecular impurity is introduced in the chamber gas, quenching of the scintillations due to collisions between the excited atoms (molecules) of the main gas and the impurity molecules takes place in addition to the photoabsorption processes. Therefore, to separate photoabsorption from collision quenching, the measurements were performed at different distances between the α-source and the chamber window, making it possible to vary the photoabsorption while keeping the quenching constant.

The measurement results are shown in Fig. 8. From the coefficient of absorption of light by the oxygen[14] we determined the effective wavelength of the scintillation light, which turned out to be 1600 ± 100 Å for both components.

D. Formation of Plasma on the Track of an Ionizing Particle

Figure 9 shows the results of measurements of the scintillation amplitude without a field and in an electric field at various pressures, in Ne, Ar, Kr, and Xe. We see that the sensitivity to the field appears in these gases at different pressures and disappears with decreasing pressure. In helium, no change of sensitivity was noted up to 20 atm.

These data confirm that gas scintillators are not sensitive to an electric field[8,9] at pressures near atmospheric. At low pressures (ionization densities), the electrons diffuse freely from the region of the track and go off to the walls of the chamber. With increasing pressure, local fields acting on the electrons by the ions increase, since the ionization density increases. It can be shown that if the gas is ionized uniformly inside a column of radius r, then we can readily obtain from the expression for the Debye screening radius[15]

$$\lambda_D / r = \sqrt{\varepsilon kT / 4e^2\eta},$$

where λ_D is the Debye-Huckel screening radius, ϵ is the dielectric constant of the gas ($\epsilon \approx 1$), k is Boltzmann's constant, e is the electron charge, η is the number of ions per unit length of track (the linear ionization density), and T is the electron temperature. Since the recombination proceeds effectively only after thermalization of the electrons[11], we put T = 300°K. It is seen from this expression that the presence of the plasma on the track of the ionized particle ($\lambda_D/r \leq 1$) is determined only by the linear ionization density η.

Using the well known ionizing ability of α-particles in various gases[16] and the values of the energy required for ion pair production[17], we can deduce from the data of Fig. 9 that the sensitivity of a gas scintillator to an electric field is observed, for all the gases investigated, at $\eta = 2 \times 10^5$ cm^{-1} (in the case of helium, the indicated linear ionization density is reached at pressures exceeding 25 atm).

The obtained linear density, in agreement with the formula presented above, corresponds to the start of formation of plasma on the particle track ($\lambda_D/r \approx 0.5$). When the plasma is formed on the track of the ionizing particle, effective recombination of the thermalizing electrons with the positive ions takes place in an ambipolar diffusion process. The electron-ion recombination is the cause of the occurrence of the field-dependent scintillation component. However, as the recombination proceeds, the ion density in the column decreases, and ultimately a time is reached when the positive space charge of the column cannot retain the electrons ($\lambda_D/r \geq 1$). The plasma decays, and the electrons go off to the chamber wall. Obviously, the fraction of the electrons that avoid recombination will continuously decrease with increasing ionization density. Indeed, as seen from Fig. 9, an increase of the gas pressure and consequently of the ionization density leads to an increase of the gas-scintillator light yield.

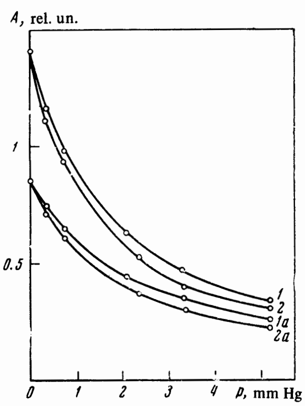

FIG. 8. Dependence of the scintillation pulse amplitude A on the oxygen-impurity pressure p. Ionization with α-particles, pressure of xenon 10 atm. Curves 1 and 1a were obtained at distance of 22 mm from the α source to the chamber window; 2 and 2a – 46 mm. Curves 1 and 2 were plotted without a field, curves 1a and 2a in the presence of an electric field ensuring a maximum decrease of the amplitude.

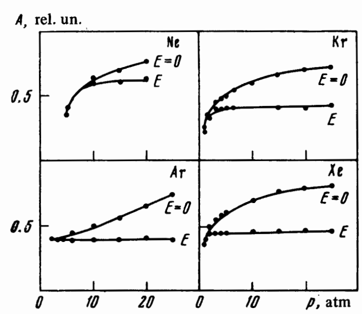

FIG. 9. Dependence of the scintillation amplitude A of various noble gases under the influence of α-particles on the gas pressure p. The curves were plotted without a field (E = 0) and in the presence of a field (E) ensuring the maximum decrease of the scintillation amplitude.

4. DISCUSSION OF RESULTS

After an ionizing particle passes through a gas, ions (X^+), electrons (e), and excited atoms (X^*) remain on the track. The main part of the scintillation light, lying in the region of the vacuum ultraviolet, can occur only when the excited atom of the noble gas goes over into the ground state. Such transitions lead to the formation of resonant photons which are absorbed in the gas. Consequently, the excited atoms cannot be the main source of the scintillation light. By colliding with the gas atoms, they form excited molecules and molecular ions, principally in the reactions

$$X^* + X + X \rightarrow X_2^* + X,$$
$$X^* + X + X \rightarrow X_2^+ + X + e.$$

The large cross sections of these reactions[18,19] lead to the formation of molecules already at a normal gas pressure within times $\tau \sim 10^{-8}$–10^{-9} sec, which is comparable or less than the time of emission of the excited atoms. The emission of the excited molecules constitutes the main part of the field-independent scintillation component. This is confirmed by our spectral measurements, which have shown that the effective wavelength of the scintillation light lies in the region occupied by the molecular continuum of xenon[20].

Ions in collisions with neutral atoms of the gas form molecular ions:

$$X^+ + 2X \rightarrow X_2^+ + X$$

within a time $\sim 10^{-8}$ sec at normal pressures[21].

Molecular ions recombine with the thermalized electrons, mainly in the reactions:

$X_2^+ + e + e \rightarrow X_2^* + e$ in a strongly ionized plasma;

$X_2^+ + e + X \rightarrow X_2^* + X$ in a dense gas;

$X_2^+ + e \rightarrow X^* + X$—dissociative recombination[15].

The excited atoms produced by dissociative recombination also form excited molecules.

Since thermalization of the electrons occurs within times $\leq 10^{-7}$ sec[22], under our conditions, the time of recombination of the thermalized electrons with the ions (as seen from Fig. 6) is $\sim 10^{-5}$ sec. The emission of the excited molecules produced upon recombination is the cause of the recombination component of the scintillation light. Since the main source of the light of both components of the scintillation is an excited molecule, it is not surprising that their measured spectral compositions are the same.

Since the total number of ions and excited atoms in the xenon irradiated by α-particles with energy 5.50 MeV does not exceed 10^6, and the number of photons in the scintillation flash is $\sim 10^4$[23], only several percent of the excited molecules emit light. The bulk of the excited molecules decays without emission. With increasing gas density, the number of quenched collisions of atoms with excited molecules (and atoms) increases, therefore the probability of light emission by the molecules should decrease. This may explain the decrease of the scintillation amplitude with increasing xenon density (Fig. 4).

The recombination efficiency depends on the ion density on the track of the ionizing particle, and consequently the yield of the scintillation light should depend not only on the energy lost by the particle in the gas but also on the ionizing ability of the particle. The higher the ionizing ability, other conditions being equal, the greater the scintillation flash. This is confirmed by the nonlinearity of the yield of scintillation light as a function of the energy absorbed in the gas in ionization by α-particles, electrons, and fission fragments[24]. On the other hand, if the ionizing ability of the particle is small ($\eta < 2 \times 10^5$ ions/cm), no plasma is produced on the particle track and there is no electron-ion recombination. In this case the light yield of a gas scintillator depends only on the loss of particle energy in the gas.

We can thus separate the following processes which determine the operating mechanism of gas scintillators:

1) Formation of excited molecules and molecular ions in collisions between ions and excited atoms with neutral atoms of gas.

2) Formation of excited molecules by recombination of electrons with molecular ions in the plasma on the track of the ionizing particle.

3) Emission of excited molecules and their de-excitation in collisions with neutral atoms.

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