

ELECTROLUMINESCENCE OF THE NOBLE GASES

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A study has been made of the characteristics of the luminescence initiated by ionizing radiation in gaseous xenon located in an electric field. The mechanism, and also some possibilities for application of noble-gas electroluminescence, are discussed.

1. INTRODUCTION

THE term electroluminescence is used to describe the emission of radiation by a material under the influence of an electric field. If a noble gas is placed in an electric field, the intensity of the light flashes due to electroluminescence initiated by an ionizing particle can be considerably greater than the scintillation flashes. References to experiments on electroluminescence initiated by ionizing particles can be found in the work of Braglia et al.^[1] (see also refs. 2-5). However, up to the present time there is no clear understanding of the mechanism of electroluminescence.

Askar'yan^[6] proposed that in strong electric fields, electron bremsstrahlung can substantially increase the amplitude of a scintillation flash initiated by an ionizing particle in the gas. We note that the process of radiation of light by electrons in scattering by neutral atoms is at present not adequately understood. For example, in the work of Firsov and Chibisov^[7] it is shown that, at least in the classical theory, in scattering of an electron by an atom the radiation of a polarized atom exceeds the contribution of the bremsstrahlung light. A calculation made by Kas'yanov and Starostin^[8] by the diagram method showed the opposite, namely, that the contribution of the atom to the radiation is two orders of magnitude smaller than the electron bremsstrahlung. However, these authors suggest that for electron energies close to the Ramsauer minimum, the contribution of atomic dipole radiation may be turned out to be comparable with the radiation of the free electron.

Szymanski and Herman^[9] have suggested that electroluminescence in strong fields can be explained by excitation of the atoms by electron impact. However, electroluminescence is observed also in weak electric fields, when there are practically no electrons with energies above the first excitation level of the gas atom.^[1] In the work of Koch^[10] it is assumed that in weak fields electrons are capable of acquiring an energy sufficient for excitation of impurities, since the excitation level of the impurity atoms can be much lower than that of the main gas. Braglia et al.^[1] assume that electroluminescence in weak fields arises from the fact that the electrons start from the track of an ionizing particle already possessing a large energy (several eV), and the conversion of the initial nonstationary electron distribution arising in the track after passage of the ionizing particle to a stationary distribution is accompanied by acceleration of some elec-

trons in the electric field to energies sufficient for excitation of gas atoms.

2. EXPERIMENTAL APPARATUS

The study of noble gas electroluminescence was carried out with the equipment described previously.^[4,11] The electroluminescence was initiated by α particles (energy 5.15 MeV) or by a pulsed x-ray beam (pulse duration 300 nsec, average photon energy 60 keV). A quartz monochromator was used to study the luminescence spectrum in the optical region. The width of the monochromator slit, while providing sufficient light intensity at the photocathode of the FEU-18A quartz-window photomultiplier used for recording, gave an accuracy in wavelength determination of 50 to 150 Å (for wavelengths from 2400 to 5600 Å). For the spectral measurements the electroluminescence was initiated by an x-ray beam from a RUT-60-20-1M equipment.

3. EXPERIMENTAL RESULTS

1. Intensity of radiation. Figure 1 shows oscillograms of the intensity of electroluminescence in xenon,

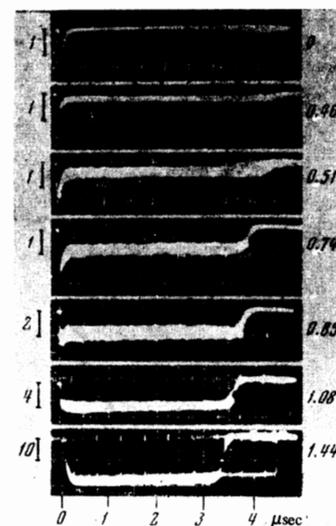


FIG. 1. Oscillograms of luminescence intensity of xenon in an electric field. On the left is the pulse-height scale in relative units, and on the right is E/P in $V/cm\text{-mm Hg}$. The xenon pressure is 25 atm. $t = 25^\circ C$. The luminescence was initiated by 5.15-MeV α particles, and the distance between the electrodes is 8.5 mm.

due to electrons from an α -particle track. In weak fields a long tail of light from electroluminescence is drawn out directly after the scintillation flash. It is apparent that the intensity of the electroluminescence falls rapidly to zero when the electrons reach the anode, and consequently the time during which electroluminescence is observed is the drift time of the electrons in the electric field from the α source, which is located at the cathode, to the anode (the range of the α particles in the gas is much less than the distance between the electrodes). At high electric field strengths the electroluminescence intensity considerably exceeds that of the scintillation flash (Fig. 1). The rise time of the electroluminescence intensity decreases with increasing electric field strength from 200 nsec at $E/p = 1$ V/cm-mm Hg to ~ 30 nsec at $E/p = 10$ V/cm-mm Hg. It is evident from Fig. 1 that the motion of the electrons to the anode is characterized by a constant intensity of electroluminescence.

2. Dependence of electroluminescence on E/p . The parameter E/p characterizes the energy variation of a charged particle moving in the gas between two collisions and, consequently, completely determines the effect of the electric field on processes involving the interaction of the charged particles with the gas atoms. In noble gases such processes include bremsstrahlung by electrons scattered by neutral gas atoms, and excitation of atoms by electron impact. If the electroluminescence is produced by these processes, the light yield of the electroluminescence (per collision) should depend only on E/p .¹⁾ If interaction of an electron occurs with several atoms at once or if the interaction of atoms with each other is important, the electroluminescence light yield should depend also on the density. Figure 2 presents in relative units the electroluminescence light yield per collision of the electron with a xenon atom, for different gas densities.

The results shown in Fig. 2 were obtained with α particles. The number of electrons pulled out of the α -particle track by the electric field was determined

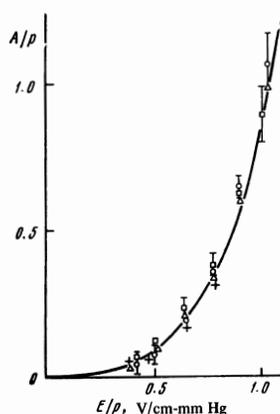


FIG. 2. Electroluminescence intensity A/p (relative units), reduced to the same number of electrons, as a function of electric field intensity E/p (V/cm-mm Hg). The electroluminescence was initiated by α particles in xenon of different densities ($t = 25^\circ\text{C}$): \square - 0.05 g/cm³, \circ - 0.125 g/cm³, Δ - 0.21 g/cm³, $+$ - 0.66 g/cm³.

¹⁾In real gases p represents the pressure which a gas of a given density would have if it were an ideal gas.

²⁾A certain decrease in the yield of ultraviolet radiation in a weak electric field is due to a reduction in the scintillation flash as the result of a decrease in the number of electron-ion recombination events in the α -particle track (see ref. 11).

from the amplitude of the ionization pulse. Figure 2 shows the probability for producing a photon in a collision of an electron with a gas atom, as a function of the electric field strength. It can be seen from the figure that the electroluminescence yield depends only on the quantity E/p and does not depend on the density ρ of the xenon, if $E/p \lesssim 1$ V/cm-mm Hg. On the other hand, for $E/p > 1$ V/cm-mm Hg, as has been shown by Szymanski and Herman,^[9] the light yield per collision depends not only on E/p but also on the gas density.

3. Electroluminescence spectrum. Figure 3 shows the light yield from xenon as a function of the electric field intensity. Here the light was detected both with a wavelength shifter which transformed the ultraviolet radiation into visible light (curve 1), and without a wavelength shifter (curve 2). It is evident from Fig. 3

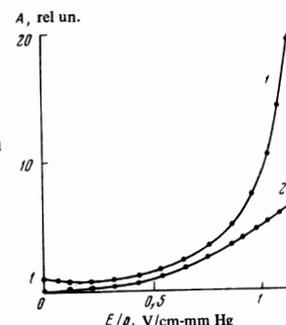


FIG. 3. Xenon luminescence amplitude A (relative units) as a function of electric field intensity E/p (V/cm-mm Hg). The electroluminescence was initiated by α particles in xenon ($p = 10$ atm, $t = 25^\circ\text{C}$). Curve 1—the entire luminescence spectrum is recorded with a wavelength shifter (quaterphenyl); curve 2—only the visible portion of the spectrum is recorded.

that for $E/p < 1$ V/cm-mm Hg the electroluminescence light lies mainly in the visible part of the spectrum. Since the radiation of xenon lies mainly in the ultraviolet region, the dominance of visible electroluminescence for small E/p indicates that in this region the electroluminescence mechanism cannot involve xenon atom excitation by electron impact. On the other hand, for $E/p \gtrsim 1$ V/cm-mm Hg (Fig. 3), the fraction of ultraviolet radiation rises rapidly.²⁾

Figure 4 shows the optical portion of the xenon electroluminescence spectrum in strong fields ($E/p \gtrsim 1$ V/cm-mm Hg). The characteristic xenon continuum is visible in the region 2000–600 Å, which is associated with quenching of excited Xe_2^* molecules (see for example ref. 12). This indicates a substantial contribution of electron-impact excitation processes in strong electric fields.

4. Effect of impurities on electroluminescence. Atomic impurities with low lying excitation levels are capable of increasing substantially the electrolumines-

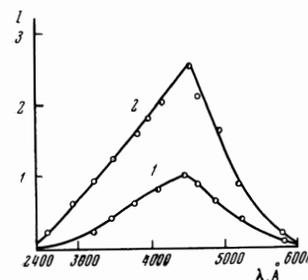


FIG. 4. Xenon electroluminescence spectrum ($\rho = 0.38$ g/cm³, $t = 25^\circ\text{C}$) for different electric field intensities E . Curve 1 - $E/p = 1.0$ V/cm-mm Hg; curve 2 - $E/p = 1.5$ V/cm-mm Hg. The ordinate is the electroluminescence intensity I in relative units.

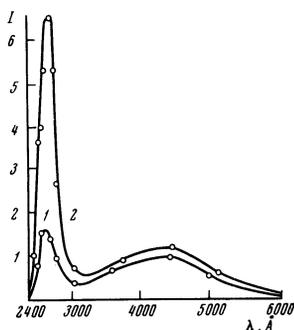


FIG. 5. Xenon electroluminescence spectrum ($\rho = 0.33 \text{ g/cm}^3$) in the presence of mercury vapor (0.002 mm Hg). Curve 1— $E/p = 1.9 \text{ V/cm-mm Hg}$; curve 2— $E/p = 2.3 \text{ V/cm-mm Hg}$.

cence light yield in the spectral region coinciding with the resonance level of the impurity.

Figure 5 shows the xenon electroluminescence spectrum in the presence of mercury vapor (0.002 mm Hg). A sharp peak characteristic of the mercury resonance line is visible. It is evident that mercury atoms effectively intercept the xenon excitation energy.

Since the excitation level of mercury lies almost a factor of two below the first excitation level of xenon, in direct excitation of the mercury atom by electron impact the mercury line can arise in weak fields insufficient for excitation of xenon atoms. However, the mercury line, as can be seen from Fig. 5, appears only in fields at which the molecular continuum of xenon is already very intense, which indicates the existence of a mechanism for transferring the energy from xenon atoms excited by electron impact to mercury atoms. Thus, in weak fields, when there are practically no excited xenon atoms ($E/p \sim 0.1 \text{ V/cm-mm Hg}$), electroluminescence cannot be explained by the excitation and quenching of impurity atoms.

Figure 6 illustrates the effect of a molecular impurity (nitrous oxide) on electroluminescence. The drop in light yield can easily be explained by the reduction of the temperature of the drifting electrons as

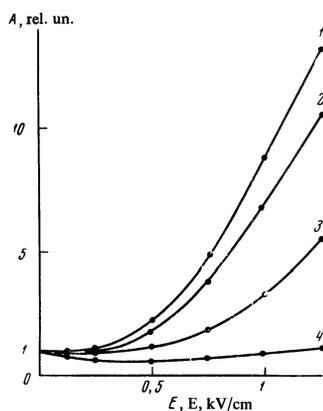


FIG. 6. Xenon electroluminescence intensity A as a function of electric field intensity E in the presence of an impurity (nitrous oxide). Curve 1—xenon without impurity ($p = 3 \text{ atm}$), 2—impurity vapor pressure $7.8 \times 10^{-3} \text{ mm Hg}$, 3—impurity vapor pressure $7.8 \times 10^{-2} \text{ mm Hg}$, 4—impurity vapor pressure $7.8 \times 10^{-1} \text{ mm Hg}$.

the result of inelastic collisions with impurity molecules. In this case the electron energy is dissipated as heat, and the electroluminescence light yield decreases.

5. Electroluminescence in pulsed electric fields. By supplying a pulsed electric field with different delays with respect to the time of passage of a 300-nsec x-ray pulse through the gas, we established that the drop in amplitude of electroluminescence with increasing delay time is produced by the decrease in the number of electrons in the chamber volume as the result of recombination.^[11] Figure 7 shows the intensity of electroluminescence as a function of delay time for a high-voltage pulse providing $E/p = 0.4 \text{ V/cm-mm Hg}$.

As can be seen from Fig. 7, electroluminescence in a weak electric field is present for pulse delays considerably greater than the thermalization time of electrons arising in the gas from absorption of the x-ray photons (in our case the thermalization time is $\sim 10 \text{ nsec}$). It follows from this that electroluminescence in weak electric fields cannot be due to acceleration in the electric field of electrons already possessing a high energy, as was suggested by Braglia et al.^[11]

4. DISCUSSION OF EXPERIMENTAL RESULTS

The experiments permit us to establish the following properties of electroluminescence: 1) The intensity of electroluminescence in a uniform field is proportional to the number of drifting electrons; 2) for an established motion of the electrons the intensity of electroluminescence is constant (see Fig. 1 and ref. 2); 3) in weak electric fields the electroluminescence light lies in the visible (and possibly infrared) region of the spectrum; 4) in strong fields, mainly ultraviolet light is emitted; 5) the ultraviolet radiation is emitted, apparently, by xenon molecules (see also ref. 2); 6) in weak electric fields the electroluminescence intensity per collision is proportional to E/p ; 7) impurities strongly reduce the intensity of electroluminescence (Fig. 6), but can increase the relative light yield in particular portions of the spectrum (Fig. 5).

The properties enumerated above permit us to draw the following conclusions regarding the mechanism of electroluminescence.

The process of noble gas atom excitation by electron impact, which is accompanied by emission of ultraviolet radiation, becomes important only in strong fields (in xenon for $E/p > 1 \text{ V/cm-mm Hg}$). Resonance photons emitted by the atoms are captured in the

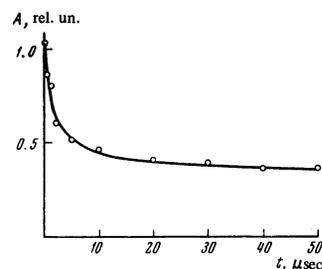
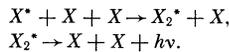


FIG. 7. Electroluminescence intensity A as a function of high-voltage pulse delay time t_d . Xenon density 0.5 g/cm^3 , $E/p = 0.4 \text{ V/cm-mm Hg}$; the height of the scintillation pulse is taken as unity.

gas, and therefore we observe mainly the ultraviolet radiation of excited noble gas atoms:



This mechanism also explains the dependence of the electroluminescence light yield in large fields not only on E/p , but also on p . In the presence of an impurity, energy transfer is possible from excited atoms (or molecules) of the main gas to the impurity. For low impurity concentrations this process is apparently considerably more probable than direct excitation of impurity atoms by electron impact (see Section 3, part 4).

In weak fields (in xenon for $E/p < 1$ V/cm-mm Hg) almost all of the light arises in elastic scattering of electrons by neutral atoms (bremsstrahlung). Only in this process can light be produced which lies almost entirely in the long-wavelength part of the spectrum. On the basis of the formulas developed by Askar'yan^[6] it is easy to estimate the number of photons from bremsstrahlung of an electron drifting in an electric field with a velocity u , emitted in a frequency interval $\Delta\omega$ per unit path along the field:

$$dN/dx \sim (\Delta\omega/h\nu) (e^2/2c^3) (\sigma n v^3/u). \quad (1)$$

Here $h\nu$ is the average photon energy, v is the electron velocity, e is the electronic charge, σ is the cross section for scattering of an electron by a gas atom, n is the number of gas atoms per unit volume, and c is the velocity of light. The same formula (except for a numerical factor close to unity) can be obtained from the results reported in refs. 8 and 13. Equation (1) is derived on the assumption that the electron energy is considerably greater than the energy of the bremsstrahlung photon, and the frequency of electronic collisions is less than the frequency of the light.

If we suppose that in xenon at a pressure of 1 atm an electric field of about 100 V/cm is produced ($E/p \sim 0.1$), then $v \sim 10^8$ cm/sec, $\sigma \sim 10^{-15}$ cm², and $u \sim 10^5$ cm/sec. In the optical region of frequencies ($\Delta\omega/\omega \sim 1/2$) under these conditions we obtain for one free electron $dN/dx \sim 0.1$ photon/cm. Here an α particle with energy of about 5 MeV in xenon produces roughly 2×10^5 electrons which, drifting in the electric field, will produce about 10^4 bremsstrahlung photons, which exceeds the intensity of the scintillation.^[14] Thus, electron bremsstrahlung is the main cause of noble-gas electroluminescence in weak electric fields.

It is obvious that the properties and mechanism presented above for electroluminescence should be valid for all noble gases.

5. SOME APPLICATIONS OF ELECTROLUMINESCENCE

Noble-gas electroluminescence initiated by charged particles is widely used for detection of ionizing radiation. Electroluminescence in very high fields is used to detect ionizing particle tracks in track-imaging spark chambers. Braglia et al.^[2] have discussed the possibility of measuring ionization density in α -particle tracks by means of electroluminescence. Varlamov

et al.^[3] have described the increase of the spectrometric capabilities of gaseous scintillators by means of electroluminescence in pulsed electric fields. We will discuss some other possible applications of electroluminescence.

1. Determination of electron drift velocity. The drift velocity of electrons in gases of any density is easily found from the duration Δt of the electroluminescence, if the electron path l in the electric field is known (see Sec. 3, part 1, Fig. 1):

$$u = l / \Delta t;$$

l must be chosen so that $\Delta t \gg \tau$, where τ is the rise time of the electroluminescence intensity. This method is suitable for use with high fields, when the electroluminescence intensity is high. Our measurements of electron drift velocity as a function of E/p in dense noble gases (up to $\rho = 0.5$ g/cm³) agree with the known data,^[15] and we will not report them.

2. Determination of the number of electrons in a gas volume. Determination of the number of electrons in a gas volume (in relative units) can be easily accomplished from the intensity of electroluminescence, since the latter is proportional to the number of electrons. If a pulsed electric field producing electroluminescence is provided at different times relative to the passage of ionizing radiation through the gas, it is possible to determine the electron recombination rate (see Fig. 7, and also ref. 11).

3. Monitoring of impurity content. As was shown in Section 3, electroluminescence depends substantially on the impurities in the gas, and consequently can be successfully used to monitor the gas purity.

The impurity content can be checked in three ways: 1) Measurement of electron drift velocity (see part 1), which is well known to depend on the impurity content in the noble gas;^[15] 2) measurement of electroluminescence light intensity (Sec. 3, Fig. 6); 3) measurement of the spectral composition of electroluminescence (Sec. 3, Fig. 5).

The last method is the most sensitive and can allow determination of certain impurities with an accuracy of 10^{-6} – $10^{-7}\%$.

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