GLOW OF GASES IRRADIATED BY SOFT X-RAYS

S. V. SAMOILOV, V. A. TSUKERMAN, and I. Sh. MODEL'

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X-ray luminescence of air, nitrogen, argon, and gas mixtures, induced by radiation of wavelength 0.3 - 1.5 A, was investigated in the pressure range from 0.01 to 760 mm Hg. It is shown that the principal role in the excitation of gas glow is played by photoelectrons and recoil electrons, produced by interactions between x-ray quanta and gas atoms. The data obtained are explained satisfactorily by combining the gas-kinetic mechanism of extinction of the excited levels with x-ray absorption in the gas. For argon and air the experimentally-determined mean light yield, under normal conditions, is 5×10^{-4} and 2×10^{-4} light photons respectively per absorbed x-ray quantum of wavelength 1.2 A, and 9×10^{-4} and 5×10^{-4} photons respectively per quantum of wavelength 0.6 A.

No x-ray luminescence of metallic surfaces proper was observed at flux intensities up to 600 r/sec. It was established that the glow observed on a metal surface in air is due to luminescence of the thin layer of gas adjacent to the surface, induced by the photoelectrons knocked out from the metal by the x-ray quanta.

INTRODUCTION

IN recent years there has been more interest in research on the glow of gases and metals under the influence of ionizing radiations. The use of gas scintillators in nuclear physics and many astrophysical problems (polar lights, glow of night sky, etc.) require a detailed study of the mechanism of glow of matter under the influence of cosmic radiation, γ rays, and fast charged particles. References 1-6 contain extensive experimental and theoretical material on the glow spectra, intensities, and durations of glows resulting from the interaction of various gases with slow and fast electrons, α particles, and positive ions. Refs. 7-11 contain the glow characteristics of noble gases used as luminophors and scintillation counters.

However, in spite of the relatively large number of investigations, the mechanism of the glow induced in gases and metals by the individual types of radiation has not been sufficiently studied. The emission of light by gases and metals under xirradiation was investigated in only two works. In 1937 Krasnikov first observed and described these phenomena qualitatively.¹² In 1955 Spicer¹³ investigated the pressure dependence of the glow inten-`sity in air, nitrogen, and argon, and also obtained the first data on the glow spectrum. The excitation mechanism for the glow of gases and metals under the influence of x-irradiation was not discussed in these works.

The aim of the present work is a more detailed

investigation of the glow of gases and metals under the influence of soft x-rays. In addition to disclosing the dependence of the glow intensity on the type and pressure of the gas, certain data have been obtained on the mechanism of transformation of xradiation into visible light.

PROCEDURE

The source of soft x-rays was a high-power dismountable close-focus type TRB-3 x-ray tube, developed by the Institute of Physical Chemistry of the Academy of Sciences, U.S.S.R.¹⁴ The tube operates at 50 kv and 40 ma. The distance between a focal center on the tungsten anode and the outer surface of the beryllium window is 20 mm. Under these conditions, the intensity of x-radiation in the window plane reaches 600 r/sec.

The chamber for the study of the luminosity of metals comprises a hermetically-sealed volume with two windows (Fig. 1a). The x-rays enter the lower beryllium window 1 and the luminous phenomena are observed through window 2. The investigated metal plate 3 is placed in the path of the x-ray beam against window 2.

The inner volume of the chamber can be evacuated to 10^{-2} mm Hg. The chamber for the study of gas glow (Fig. 1b) was made taller to exclude the additional illumination that might arise on the upper end of the chamber. To absorb the undesirable glow from the sidewalls, the latter were covered with black paper. In spectral investigations, win-



Fig. 1. Chambers for the investigation of x-ray luminescence: a - of metals and b - of gases. 1 - beryllium window; 2 - glass window; 3 - investigated metal; 4 - recording instrument.

dow 2 was replaced by a plate of transparent quartz 3 mm thick. The total thickness of the beryllium placed between the anode of the tube and the inner cavity of the chambers was 2.3 mm.

The luminous phenomena in the chambers were observed with the aid of a photomultiplier, a photographic camera, and a spectrograph. The first method made it possible to evaluate quantitatively the intensity of the glow as a function of the type and pressure of the gas and of other characteristics. All experiments were carried out with type FEU-19 photomultipliers, whose output signals were recorded with an EO-4 oscillograph. The photomultiplier sensitivity was 700 - 1200 amp/lu. The oscillograph gain was $10^2 - 10^5$. At maximum gain, a 1-mm deflection of the beam on the oscillograph screen corresponded to a flux of 2×10^{-11} lu on the photocathode.

The photographic camera was used to determine the spatial distribution of the glow intensity within the volume of the chamber. The relative aperture of the objective was 1:1.5, the focal distance was f = 50 mm. Using type D panchromatic motion picture film (250 - 350 GOST units), the glow could be photographed in the chambers at exposures from 5 to 200 minutes.

The spectral investigations were made with a type Q-12 Zeiss quartz spectrograph. The high intensity of the x-rays source made is possible to record glow spectra with photographic plates, without resorting to photomultipliers, as Spicer had to do.

EXPERIMENTAL RESULTS

Figure 2 shows the results of the first measurements of glow intensity as functions of the air pressure for Be, Cu, Mo, Sn, and Pt. The measurements were made with a photomultiplier using the circuit of Fig. 1a. As can be seen from the graph, the absolute light yield increases with increasing



Fig. 2. Dependence of glow intensity on the air pressure in the chamber, for various metals.

atomic number of the metal. As the pressure is reduced from 760 to 7 - 10 mm Hg, most metals exhibit an increase in glow intensity. Further reduction in pressure leads to a monotonic decrease in intensity. Finally, at approximately 10^{-2} mm Hg and below, the photomultiplier detects no noticeable light yield even at maximum gain. These unexpected results show that the observed glow is not connected with fluorescence induced in metals by the x-rays, as was proposed in Ref. 12. It has been suggested that the glow of gas in the chamber is excited by electrons, knocked out by the x-ray quanta from the metallic surface and from the gas atoms as the result of the photoeffect. Figure 3 shows photographs of the glow of the air in the chamber, confirming this suggestion. At a relatively high pressure (76 mm Hg) one observes, in addition to the glowing column of air in the lower portion of the chamber at the outlet window, also



Fig. 3. Photographs of x-ray luminescence of air in the chamber of Fig. 1a: a - pressure 76 mm Hg; b - pressure 7 mm Hg; c - experimental setup for the confirmation of the electronic nature of the observed glow;<math>d - photograph of x-ray luminescence at 7 mm Hg, using the scheme of Fig. 3c.

a bright glow of the surface of the plate (marked by an arrow in Fig. 3a). Such a photograph can readily lead to the erroneous conclusion that the metal surface fluoresces. However, the next photograph, made at 7 mm Hg, contradicts such a conclusion. Here the glowing region near the surface broadens noticeably, and merges with the glowing cone in the lower portion (Fig. 3b). Upon slight modification of the experiment, this phenomenon manifested itself even more clearly. The platinum plate was rotated in the chamber in such a way, that the objective of the camera did not "see" the surface facing the source of x-rays (Fig. 3c). In spite of this, the pressure of the glow of the air in the chamber turned out to be analogous to that of the preceding photographs (Fig. 3d).

Further confirmation of the electronic nature of the excitation of the glow of the gas under xirradiation, and also data on the energy of the electrons that excite the glow, were obtained by photographing the glow of the air in a magnetic field. The setup of the experiment is clear from Fig. 4a. A hole 10 mm in diameter was drilled through the lower pole of a horseshoe shaped electromagnet to



Fig. 4. X-ray luminescence of air in magnetic field. a - experimental setup, b - without magnetic field, c - with magnetic field.

permit passage of the x-rays through the gap between the poles. To increase the number of secondary electrons, the surface of the upper pole opposite the source of radiation was covered with a lead plate. With electromagnet coils energized, the field intensity in the gap between the pole is ~ 300 oersted. Photographs of the glow of the air in the gap between the poles without and with a magnetic field are shown in Figs. 4b and 4c respectively. It is seen quite distinctly that in the presence of a magnetic field the glowing region contracts towards the axis of the polepieces and assumes the shape of a glowing column. An estimate shows that the electron energy does not exceed 1000 v in a field of 300 oersted.

The pressure dependence of the intensity of the glow of air and argon, in the absence of a metallic surface in the chamber, is shown in Fig. 5 (solid curves). The experiments were made in the cham-



Fig. 5. Dependence of the intensity of x-ray luminescence of gas on the pressure. Solid – authors' data, dotted – Spicer's data.(Ref. 13).

ber shown in Fig. 1b. For convenience in comparison of the curves among themselves and with the results of other investigators, the ordinates represent the glow intensity in percent of the intensity at 760 mm Hg. The course of the curves is seen to be similar to that of the curves of Fig. 2. Up to pressures of 100 - 200 mm Hg for air and 300 - 400mm Hg for argon, the glow intensity remains practically unchanged. Further reduction in pressure is accompanied by a monotonic decrease in intensity. The increased glow intensity in the 5-50mm Hg region, observed in the experiments with the metallic plates, is not seen in this series of measurements. An analogous behavior of the curves was established in experiments with helium, oxygen, and nitrogen. The general character of the observed laws is in close correspondence with the pressure dependence of the glow as obtained by Spicer¹³ for air (dotted curve of Fig. 5). The absolute glow intensity depends on the type of gas. At atmospheric pressure, the glow intensities of helium, oxygen, air, nitrogen, and argon are related as 1:2:10:35:1,000. The glow intensity of a gas mixture is determined by the composition and the ratio of the mixture components and by the pressure. Figure 6 shows graphs for the glow intensity of mixtures of argon with oxygen (a) and of argon with nitrogen (b). The abscissas represent the percentage of oxygen or nitrogen constant in the argon, while the ordinates represent the glow intensity in percentage of the intensity of pure argon. Three series of measurement were made for pressures p = 50, 150, and 350 mm. In all cases, a small admixture of oxygen (up to 1-2%) has reduced the glow intensity by a factor of several times. The "quenching" influence of the oxygen is stronger at high pressure than at low pressure. The glow of an argon-nitrogen mixture has a different character. Here a small addition (1-2%) of nitrogen increases the glow intensity. Further in-

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crease of the nitrogen concentration results in a slow decrease in intensity.

Figure 7 shows the pressure dependence of the glow intensity of a mixture of 80% argon plus 20% oxygen. For comparison, the same graph shows the pressure dependence of the glow intensity of air. The character of the two curves is practically



Fig. 7. Dependence of intensity of x-ray luminescence of air (a) and mixture 80% Ar + 20% O_2 (b) on the pressure.

identical. It is interesting to note that the absolute intensity of the glow of a mixture of argon with oxygen is only slightly higher than the glow intensity of air at equal pressures. This experiment refutes Spicer's point of view concerning the influence of the natural-argon content of air on the intensity of its glow.

An attempt was made to establish the connection between the glow intensity of gas and the hardness of the x-rays. For this purpose, the soft component of the x-ray spectrum was "cutoff" with the aid of aluminum filters 0.05 to 2.3 mm thick. The drop in glow intensity of air and argon had an exponential dependence on the thickness of the filter, with a variable exponent. From the resultant curves one can conclude that soft x-rays produce a more effective excitation. It must be noted that our experiments did not disclose the linear dependence of the logarithm of glow intensity on the filter thickness, as indicated by Spicer.

Figure 8 shows the result of microphotometry of a spectrogram of argon glow at atmospheric pressure. In the investigated spectral range, 6,000

Fig. 6. Dependence of intensity of x-ray luminescence of argon on the concentration: a - of oxygen, b - of nitrogen, for pressures $p_1 = 50$, $p_2 = 150$, and $p_3 = 350$ mm Hg.

-2,500 A, seven lines were observed in the violet regions. The positions and relative intensities of these lines are in good agreement with the lines of the neutral atoms in the spectrum of argon excited by an electric discharge (shown by vertical bars).¹⁵ The most intense lines were those at 3390, 3600, 3840, 3180, and 4180 A.



Fig. 8. Spectrum of x-ray luminescence of argon at atmospheric pressure (the lower lines are those of argon in gas discharge, coinciding with the x-ray-luminescence lines).

The data obtained on the x-ray spectrum, intensity, and spectral composition of the visible glow excited by x-rays has made it possible to estimate approximately the yield of the light photons per x-ray quantum. Such a calculation was made for argon and air at atmospheric pressure. The center line of the wavelength of the light radiation was taken to be 3500 A. The calculation results are listed in the table.

Light yield of x-ray luminescence (p = 1 atmos, t = 20°C)

Gas	wavelength of x-rays, A	light yield per absorbed x-ray quantum in photons, λ = 3500 A
Air Argon	0.6 1.2 0.6 1.2	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$

Errors in the determination of the spectra of the x-ray and optical radiations, inaccurate knowledge of the spectral characteristics of the photocathode of the multiplier, errors in the account for the geometry of the experiment, and several other factors difficult to control may cause these values to deviate from the true values by a factor of 4-5. These data must therefore be considered only as tentative, illustrating from the quantitative point of view the effective cross-section of x-ray luminescence in gases.

EVALUATION OF RESULTS

Let us consider the possible mechanism for the transformation of an x-ray radiation into visible light on a metallic surface. Ginzburg and Frank have described the occurrence of transition glow upon passage of fast charged particles through the separation boundary between vacuum and a metal.¹⁶ However, to obtain a noticeable glow of a metal under the influence of electrons, their energy must be not less than several tens of ky. It is known that the energy of most photoelectrons "knocked out" from a metal by soft x-rays does not exceed 1 kv. At such energies, the probability of glow due to the transition effect is vanishingly small. It is possible to represent the occurrence of visible glow on a metallic surface as being due to the long-wave region of the bremsstrahlung spectrum. In this case the radiation of light can take place only in very thin surface layers of the metal, on the order of 10^{-6} cm. Even for soft radiation, the number of x-ray quanta converted into light in such a thin layer is negligible. The absence of a noticeable glow of metallic surfaces in vacuum confirms these qualitative considerations.

With the photomultiplier sensitivity having a maximum of 10^{-11} lu, we can state in any case that the yield of light photons from the metallic surface per x-ray quantum is less than 10^{-8} , and is at least two orders of magnitude less than the glow intensity of air, as recorded in our experiments. The apparent glow of a metallic surface in air must thus be attributed to glow of the layer of gas adjacent to the surface, induced by the electrons knocked out by x-ray quanta from the metal. Since the mean energy of such electrons is 1,000 ev, their range in air amounts to ~ 2.5×10^{-2} cm. A glowing layer of gas of such thickness is perceived by the observer as a glowing metallic surface.

The increase in the absolute intensity of glow with increasing Z of the metal, as indicated in Fig. 2, does not contradict this statement. The glow of the photoeffect increases with increasing Z, and the number of secondary electrons exciting the gas atoms should be greater for a heavier than for a lighter metal. There are no grounds for assuming that the mechanism of glow excitation in gas differs from the above in the absence of a metal.

An estimate of the probability of direct excitation of the levels of the argon atoms by 1.2-A x-ray quanta shows that this probability is at least 10^7 times as small than the probability of excitation of the levels of the atoms by electrons "knocked out" by the photoeffect or by recoil electrons. The observed laws of glow of gases, excited by fast electrons and other charged particles, confirm this opinion. The pressure dependence of the glow intensity obtained by Grün and Schopper⁴ for nitrogen and for an argon-nitrogen mixture by irradiating these gases with α particles from polonium, is in good agreement with the laws of x-ray luminescence of gases.

The spectra of the x-ray luminescence of gases also confirm the principal role of fast electrons in the excitation of the glow. Fan has shown¹ that as the electron energy increases, the spectrum of the gas glow shifts towards the short-wave region. It is known that the most intense argon-discharge spectral lines, when the energy of the electrons does not exceed 100 v, are located in the 8100 – 8500 A and 4700 - 3900 A ranges. In our case, the most intense spectral lines lie between 3400 and 3800 A.

Let us consider in greater detail the effect of the pressure and of the composition of a gas mixture on the intensity of the gas glow. Spicer proposes that the pressure dependence of the glow intensity is determined by the change in the rate of electron-ion recombination with pressure. This assumption is not corroborated by calculations or by experimental data. The probability of direct recombination at negligibly small electron and ion concentrations (not more than 10^{12} ions per cm³) is very small. In addition, the recombination radiation should have a continuous spectrum. Our and Spicer's experiments resulted only in line spectra. The experimental relations are explained much better by the gas-kinetic mechanism. It is known that the average lifetimes of the excited levels can vary for different gases over very wide range, from 10^{-2} to 10^{-10} sec. The probability of transitions from the excited levels with radiation of light photons increases with diminishing lifetime. For argon atoms, the fundamental levels with light radiation have average lifetimes of $10^{-7} - 10^{-9}$ sec and in addition, there exist metastable levels with a lifetime $\sim 10^{-2}$ sec. The average time interval

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between collisions of argon atoms amounts to 1.64×10^{-10} sec at p = 1 atmos and 1.2×10^{-7} sec at p = 1 mm Hg (t = 20°C). Comparing these quantities, it is easy to note that, at pressures close to atmospheric, the time between two collisions of argon atoms is 1-2 orders shorter than the glow time of the ground levels. The glow intensity under such conditions, for a given number of excited argon atoms, will be less than the glow intensity for the same number of excited atoms at a pressure ~1 mm Hg, when the time between two atom collisions exceeds the lifetime of the excited level.

The number of excited levels n, which glow at a given pressure p, is expressed by a function of the type

$$n = N (1 - \exp\{-t_0 k / \tau_0 p\}), \tag{1}$$

where N is the total number of excited levels, τ_0 is the average lifetime of the excited level, t_0 is the average time between two collisions at p = 1 atmos, and k is a coefficient that accounts for the number of collisions per extinction event (ratio of gas-kinetic cross-section to the extinction cross-section). On the other hand, reducing the pressure reduces the x-ray absorption and correspondingly the number of excited levels per unit volume. This dependence is of the form

$$N \sim I_0 (1 - \exp\{-\mu x p\}).$$
 (2)

Here I_0 is the intensity of the x-rays, μ the linear coefficient of attenuation of the radiation in the gas at a pressure of 1 atmos, and x is the thickness of the gas layer absorbing the radiation. The interaction of these two opposing processes is shown in Fig. 9. Curve 1 shows the gas-kinetic extinction as a function of the pressure, in accordance with expression (1), while curve 2 shows the dependence of the absorption on the gas pressure (2). The superposition of the two laws is represented by curve 3:

$$n \sim I_0 (1 - \exp\{-\mu x p\}) (1 - \exp\{-t_0 k / \tau_0 p\}).$$
 (3)

This curve has an extended horizontal section in a relatively large pressure range, in good agreement with the experimental data.

The mechanism described makes it also possible to explain the experimentally-observed increase in glow intensity in the presence of a metallic surface at pressures from 1 to 10 mm Hg (curves of Fig. 2). In contradistinction with x-ray luminescence of a pure gas, the number of secondary electrons knocked out here from the surface of the metal does not diminish with decreasing pressure, and stays at approximately the same level. Thus, in a comparatively wide pressure region, the principal role is played here by gas-kinetic extinction, and the total intensity recorded by the photomultiplier should increase with diminishing pressure. Only after the mean free path of the electron in the gas begins to exceed the dimensions of the observed glowing region does a noticeable decrease in glow intensity take place. Naturally, the quantitative relations here are determined to a great extent by the geometry of the experiment. However, a numerical estimate of the results of the graph of Fig. 2 confirms these considerations quite well.

The observed laws of the glow of gas mixtures are also in good agreement with the mechanism of gas-kinetic extinction. It is known that a small addition of oxygen exerts a considerable quenching influence on the glow of a gas discharge.¹⁷ Oxygen is characterized by transitions from excited levels without radiation of light.³ The large number of energy levels in this diatomic gas leads to a practically resonant "selection" of the energies of the excited levels of argon, causing an extinction of the x-ray luminescence even at small oxygen concentrations. At low pressures of argon-oxygen mixtures, the collisions between the atoms of these gases are less frequent and the quenching influence of the oxygen is less pronounced (see Fig. 6a). This process, whereby the oxygen extinguishes the excited levels of gas atoms, is apparently the decisive factor not only for argon, but also for air. A similar course of curves for the mixtures 80% Ar + 20% O_2 and $N_2 + O_2$ (air), shown in Fig. 7, confirms the above.

To the contrary, a small addition of nitrogen increases the intensity of argon glow (Fig. 6b). An analogous phenomenon was observed by Grün and Schopper,⁴ where, at nitrogen concentrations in argon from 1 to 15%, an increase in glow intensity by 5-7 times compared with the glow intensity of pure argon was observed under the influence of α particles. The spectral investigations of the glow of argon under the influence of charged particles, performed by Bennet and Hughes,² have shown that a slight concentration of nitrogen in argon (0.001%) is sufficient to excite intensely the lines of the second positive group of neutral molecules of N_2 . The excitation potential of the highest level of this group corresponds to the potential of the metastable state of the argon. Thus, unlike in the case of oxygen, the interaction between the excited atoms of argon with the nitrogen atoms leads to an increase in the glow intensity owing to the radiation of the metastable levels.

It is interesting to note that the laws for the glow of gases and gas mixtures under the influence



Fig. 9. Dependence of gas-kinetic extinction (1), radiation absorption (2), and total light yield of x-ray luminescence (3) on the gas pressure: a - argon, b - air.

(For portions of curves 1A and 2A, the vertical scale has been increased by 100 times.)

of ionizing radiations depends relatively little on the nature of the radiation. Fast electrons, x-rays, α particles, and other energetic radiations excite approximately equal spectra. Fan, who studied the glow spectrum of air excited by electrons, protons, and helium ions has shown that different charged particles of equal velocity, excite very similar spectra. The dependences of the glow intensity on the pressures and concentrations of mixtures of various gases turn out to be exactly similar, and show little dependence on the type of ionizing radiation.

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One can consider it established that at ionizingradiation energies that exceed by many times the ionization potential of the gas, the glow is due to the electronic transitions and is determined essentially by the atomic and molecular properties of the the gas, by its density, and by its impurities.

In conclusion, we shall mention several practical consequences of the above experiments, which are of certain interest to workers with gas scintillators. An analysis of the curves of Fig. 9 shows that a considerable increase in pressure in a gas scintillator does not lead to an increase in the lumines-cence intensity (in the case of registration of charged particles it may even lead to a reduction in the glow intensity). Calculations show that at a pressure of ~ 3 atmos the intensity of lumines-cence, excited in argon by 1-A x-rays, begins to diminish.

When the chamber is filled with a gas scintillator, it is necessary to rid the gas thoroughly of all possible impurities, which may extinguish the glow — oxygen, carbon dioxide, nitrogen oxides, etc.

It is also possible to propose the following method for increasing considerably the light output of a gas scintillator for x- and γ -rays. It is known that the ordinary gas scintillators have little sensitivity to radiations of this type. If a system of plane-parallel metal foils is placed in the scintillation chamber perpendicular to the plane of the photomultiplier cathode, then the electrons knocked out by the x- or γ -rays from the surface of the foils will produce an intense glow of the gas layers located in the gaps between the foils. By choosing suitable foil thicknesses and materials and gap thicknesses, and by choosing the kind and pressure of the gas, it is possible to increase the light yield of such a gas scintillator by many times, leaving unchanged the remaining qualitative indices, the low glow time, and the simplicity of construction.

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