INFRARED EMISSION INDUCED BY ELECTRONS MOVING IN NOBLE GASES AND LIQUIDS

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We report the first experimental results of infrared emission induced by the passage of ionizing particles through moderately dense noble gases. A rich phenomenology has been revealed, which is independent on the nature of the incident particles (protons or electrons). Two different behaviors of the IR emission have been detected. The first one is believed to be induced by thermalization processes of secondary electrons in absence of an externally applied electric field and it seems to be related to scintillation phenomena in the UV range. The second one is the IR emission observed when thermalized electrons are drifted in the gas under the influence of an electric field.

Introduction

A very important issue in high energy physics is the development of efficient detectors capable of tracking the energy released by an energetic particle passing through matter.

Among the most widespread detectors are UV scintillators, which exploit the light emission produced by the deexcitation of optically excited states generated by the ionizing particles crossing the detector medium [1].

Rare gases are very often used as scintillating media. Since their scintillation light lies in the VUV band, there are several reasons, technical as well as physical, to search for emission mechanisms in a region at much larger wavelenghts λ. In fact, the collection efficiency of light guides is better in the visible and IR region. The number of photons emitted per unit amount of energy released by the particle in the gas is proportional to λ. Finally, incident particle-matter interaction processes at small transferred energy E are more likely because the energy transfer cross section increases strongly with decreasing E.

A further puzzling topic is that the W-value (the mean energy necessary to an ionizing particle to produce an electron-ion pair in the gas) is higher than the ionization energy I of the isolated atom [1]. Therefore, it is reasonable to raise the question if the missing energy $W - I$ might be dissipated by exciting low-lying levels of the system. In this case, further thermalization might result in IR emission.

It has also to be pointed out that IR emission has been foreseen for the rare gases Ar and Xe [2, 3], but there is no clear experimental evidence until now, although some atomic lines in the near IR region have been observed [4]. For these reasons we have decided to investigate the emission of rare gases in the IR band induced by ionizing particles.

Experimental Apparatus

The experimental technique is very simple. An ionizing particle beam enters an ionization chamber containing the gas under investigation. The chamber can be filled with gas up to 3 MPa. The emitted light exits the cell either through a quartz or sapphire IR window. If necessary, suitable UV filters can be inserted after the IR window. When recording light spectra a Bruker Equinox 55 Michelson spectrometer can be inserted in the optical path to the sensor.

A commercial InGaAs photodiode detects the IR light emitted during the passage of the particle through the gas. The photodiode quantum efficiency is QE>80 % in the λ-range 0.95 - 1.55 μm. The photodiode signal is detected by a charge amplifier and recorded by a digital oscilloscope.

A simplified schematics of the experimental setup is shown in Fig. 1.

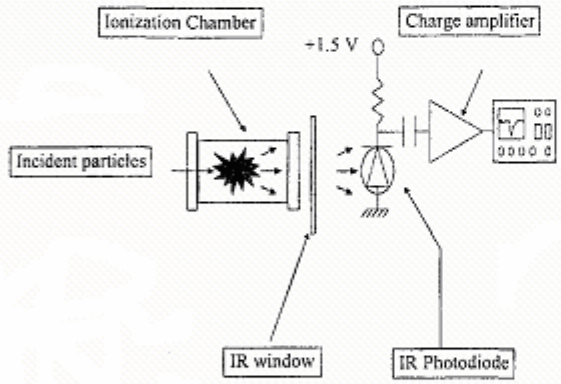

Fig. 1. Schematics of the experimental setup.

Measurements are carried out by using either protons or electrons. In the former case, the gas is excited by a 5 MeV, 1 nA current, proton beam extracted from the van de Graaf accelerator of the INFN-LNL Laboratories. The beam is chopped into bunches 50 to 400 μs long. The proton beam enters the cell through a 20 μm thick iron window. In the electron case, a home-made electron gun produces a 70 keV beam entering the cell through an 8 μm thick Kapton window. The electron bunch has a duration of ≈ 35 ns and contains approximately 1 nC charge.

An electrode setup consisting of a wire anode assembly can be mounted into the cell in order to measure the light emission during the drift of the secondary electrons.

Experimental Results

The IR emission occurs when the particle beam produces primary ionization and excitation. If an electric field is applied in order to drift the secondary electrons produced, a further increase of the intensity of the emitted light is observed. In Fig. 2 we show a typical IR signal obtained in Ar gas (P=0.1 MPa, room temperature) by using a broader band sensor (HgCdTe, bandwidth 1-12 μm) and by ionizing the gas with the proton beam [5]. In the upper part of the figure, the temporal evolution of the IR light is shown in absence of an applied electric field. The time behavior of the light pulse is closely correlated to the shape of the proton bunch. Obviously, no secondary electrons are collected at the anode.

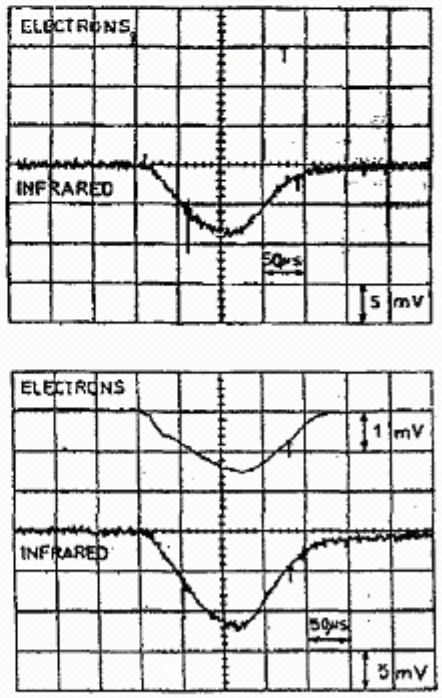

Fig. 2. IR signals in Ar gas. Top: without electric field. Bottom: with electric field applied.

In the lower part, a similar signal is recorded when an electric field is applied. In

this case, secondary electrons are collected at the anode and the light intensity appears to have increased by approximately 30 %.

Thereafter, we have to distinguish two different emission mechanisms. The first one is related to thermalization processes of secondary electrons and is called *prompt* emission. The same behavior is observed also with the electron beam.

The second one seems to be induced by the (slow) drift motion of electrons in the gas and we call it *drift* emission.

Prompt Emission

A typical IR proton-beam-excited spectrum obtained in Ar is shown in Fig. 3. Two atomic lines are observed in the investigated λ–range. The same results are obtained by exciting the gas with the electron beam. We tentatively attribute the strongest line to the atomic transition 4s'$[1/2]^0$ - 4p[5/2] of Ar I. The weakest one might be attributed to the 4d' ^{2}D – (^{1}D)4f$[2]^0$ transition of Ar II. It is not yet clear why only these two lines are observed in a region where a plenty of atomic transitions are known to occur [6]. A similar phenomenology has been observed by other authors in the near IR spectra of Ar and other noble gases and has been explained by assuming that the beam excitation mechanism is very different from the excitation mechanisms in various discharge sources [4].

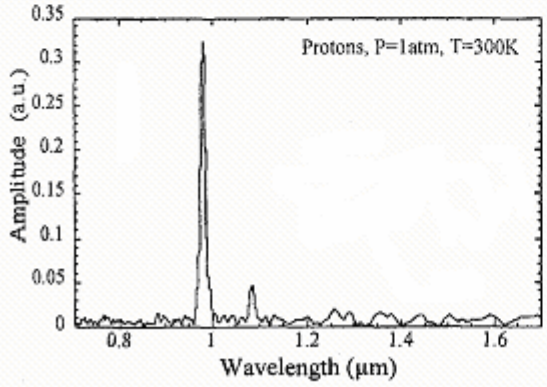

Fig. 3 Prompt proton-beam induced emission spectrum in Ar gas at P=0.1 MPa and room temperature.

By increasing the gas pressure up to P=3.0 MPa, the recorded spectra for Ar do not change significantly. The spectrum features in Ne are very similar to those in Ar, without any pressure dependences.

The situation in Xe gas is radically different. Even at the lowest pressure investigated, P ≈ 0.1 MPa, the beam-excited spectrum of Xe is overwhelmed by a very broad structure, which might obscure atomic features similar to those observed in Ar or Ne. In Fig. 4 we show the spectrum recorded in Xe gas at P = 0.3 MPa and at room temperature. Again, no differences are observed when different exciting beams are used. The presence of a broad continuum band could be explained as the consequence of radiative decay of highly excited states of diatomic molecules (excimers) to lower lying levels.

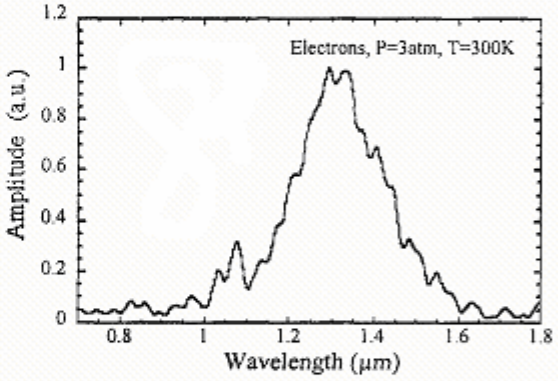

Fig. 4 Prompt e-beam induced emission spectrum in Xe gas at P=0.3 MPa and room temperature.

The previously observed VUV scintillation in high-pressure Xe and Ar is essentially caused by the same mechanism [3]. In the VUV case, there is a radiative decay from excimer levels to the repulsive molecular ground state of the normally monoatomic gas [3]. The difference between the IR and VUV scintillation is that in the IR case the transitions do not involve the repulsive molecular ground state as in the VUV case. They rather occur between excited levels. A schematic diagram of some possible transitions leading to IR and VUV scintillation is reported in Fig. 5 [3,7].

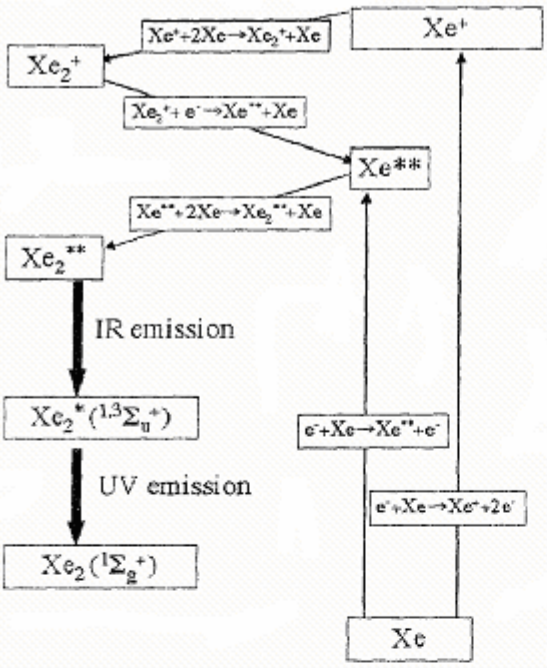

Fig. 5 Schematic diagram of the energy levels of the Xe_2 excimer leading to IR and VUV scintillation.

The mechanism to populate the excimer levels is essentially a sequence of collisional events and is possible only at relatively high densities when three-body collisions are not negligible. The incident particle (e.g., an electron) excites or ionizes the gas according to the following reactions [7]

$$e^- + Xe \rightarrow Xe^+ + 2e^-$$
$$e^- + Xe \rightarrow Xe^{**} + e^-$$

Three-body collisions give origin to association and formation of the molecular ion Xe_2^+

$$Xe^+ + 2Xe \rightarrow Xe_2^+ + Xe$$

A dissociative recombination reaction gives origin to an excited neutral in a dissociating state according to the reaction

$$Xe_2^+ + e^- \rightarrow Xe_2 \rightarrow Xe^{**} + Xe$$

Finally, again three-body association reactions produce eventually bound molecular levels according to the scheme

$$Xe^{**} + 2Xe \rightarrow Xe_2^{**} + Xe$$

The IR scintillation may be the result of the radiative decay of a Xe_2^{**} excimer to another Xe_2^* excimer of lower energy in the reaction

$$Xe_2^{**} \rightarrow Xe_2^*(^{1,3}\Sigma_u^+) + \gamma(IR)$$

and the VUV scintillation is due to the transition between the $^{1,3}\Sigma^+_u$ and the $^1\Sigma^+_g$ states of the excimer

$$Xe_2^*(^{1,3}\Sigma_u^+) \rightarrow Xe_2^*(^1\Sigma_g^+) + \gamma(VUV)$$

This reaction scheme for the IR emission has been suggested by Mulliken [2]. According to his calculations, the IR emission should occur around 2 μm in Xe. He also predicts the excimer continuum band at ≈ 1 μm for Ar, but neither we nor other researchers [3] did observe it.

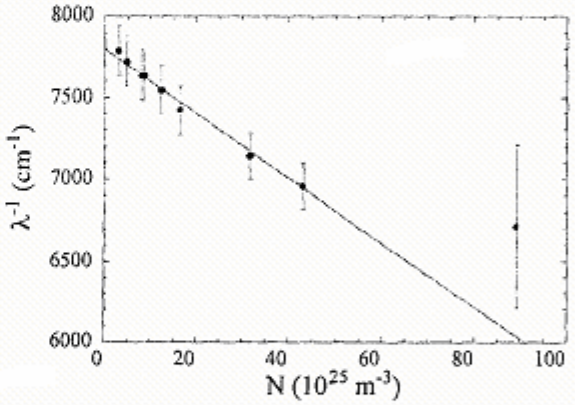

Fig. 6 Red shift of the center of the emission peak in Xe gas as a function of the density. The meaning of the line is explained in the text.

A very interesting feature of the Xe spectra is that the centroid of the continuum band (see Fig. 4) shifts to larger wavelengths as the pressure is increased. In Fig. 6 we show how the peak position, expressed in units of energy, shifts as the gas density is increased. The gas pressure ranges between ≈ 0.1 MPa and 3 MPa. The density is computed using the NIST equation of state [8].

The red shift observed is practically linear with the gas density. The experimental point at the highest density slightly deviates from linearity, probably because its wave-

length lies very close to the IR cutoff of the photodiode and this fact produces a distortion of the continuum structure of the spectrum.

Other authors [3] claim that a similar pressure induced red shift of the VUV scintillation have been detected in the same pressure range. In any case, however, the observed maximum red shift in the VUV case amounts only to nearly 1 % and is moreover comparable to the experimental error. Furthermore, more precise and recent measurements of heavy-ion excitation of rare gas excimers at lower pressures did not detect any red shift in the same VUV range [9].

In addition to the red shift of the IR spectrum centroid, we noted also a broadening of the spectrum itself, as shown in Fig. 7.

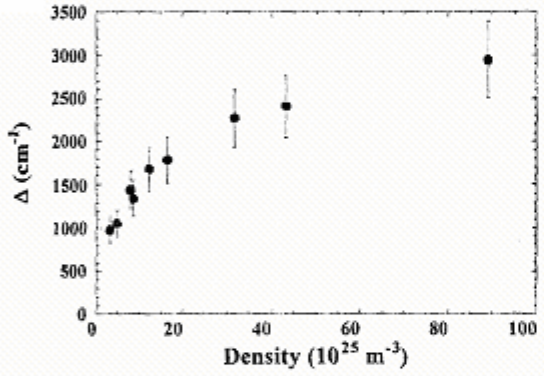

Fig. 7. Pressure broadening of the Xe emission spectrum.

Owing to the large densities involved, it is reasonable to argue that the nearly linear increase of the spectrum width with increasing density is due to collisional events [10]. It also interesting to note that the relative width $\Delta \equiv \Delta(1/\lambda)$ of the IR peak reported here and that of the VUV peak [3] are comparable. This means that the different excimer states involved in the VUV and IR transitions have nearly the same lifetime.

However, in the VUV emission, a narrowing of the line with increasing pressure is observed, in contrast with the present results. This fact has been qualitatively explained as a consequence of the absorption in a ground-state population of the unexcited Xe_2 molecule that increases with increasing pressure. This mechanism is claimed to be responsible for the pressure induced red-shift, too [3].

We suggest that different physical mechanisms should explain the observed IR continuum red shift. The polarization of the Xe atoms surrounding the excimer could in fact decrease the Coulomb interaction between the excited electrons of the Xe_2^* and the ion-core, in analogy to what happens for Mott-Wannier excitons in solids [11]. According to this model, a linear dependence of the transition energy as a funcion of density can be easily derived. The wavelength λ of the IR continuum centroid can be written as

$$\frac{hc}{\lambda} = R\left(\frac{1}{K^2 n_f^2} - \frac{1}{K^2 n_i^2}\right) \qquad (1)$$

where R is a Rydberg-like constant, n_f and n_i are the "principal quantum numbers" of the final and initial states. K is the dielectric constant of the gas and at the low density of the experiment it can be safely approximated by

$$\frac{1}{K^2} = 1 - 2N\alpha \qquad (2)$$

where N is the gas number density and α is the atomic polarizability of Xe ($\alpha = 16\pi\ 10^{-30}$ m^3). By assuming that R, n_f, and n_i are constant, and inserting Eq. (2) into Eq. (1), we get for the central wavelength

$$\frac{1}{\lambda} = A(1 - 2N\alpha) \qquad (3)$$

where A is a suitable constant.

Moreover, since the excimer are found in highly-excited, Rydberg-like states, the outer electron is in an orbit of very large radius and the electron wave function spans over several atoms at once. Therefore, many-body effects, first introduced by Fermi [12], must be considered, which give rise to a shift V_0 of the total electron energy, called *Fermi shift*, proportional to the gas density

$$V_0 = \frac{2\pi\hbar^2 a}{m} N \qquad (4)$$

where a(≈ -3.09 Å [13]) is the scatter length of the electron-atom interaction cr section. This contribution must be added i Eq. (3) thus obtaining

$$\frac{1}{\lambda} = A - CN$$

where $C = 2A\alpha - \hbar a / mc$ and A must be ted to the experimental data. In Fig. 6 the l is Eq. (5) where A (≈ 7800 cm^{-1}) is the o adjustable parameter. It is very important realize that both contributions to the ene shift (polarization and many-body effe have to be considered simultaneously. If neglects V_0, the slope $2A\alpha$ turns out to b 2.5 times smaller than the actual slope, if true atomic polarizability is used, because zero density intercept does not practically change. On the contrary, if the polarization mechanism is neglected, the resulting slope amounts only to ≈ 60 % of the actual one.

Therefore, we believe that there is evidence that the observed IR spectra in Xe are produced by transitions between Rydberg states of Xe_2 excimers.

We have also carried out preliminary measurements in liquid Ar and Xe [14,15]. There is evidence of IR scintillation in both liquids, although the light output is much lower than in the gas. Moreover, it seems that the spectra in the liquids differ also qualitatively from those in the gases.

Drift Emission

We have also carried out measurements of integrated scintillation light output in gaseous Ar and Xe, and in their mixtures using a cell with a built-in electrode assembly in order to drift the secondary electrons under the influence of an external electric field [5]. The wavelength band over which the light is integrated is that of the InGaAs sensor.

In Fig. 8 we show the dependence of the integrated light signal (normalized to the ionization charge collected) as a function of the high voltage applied to the electrode system.

Main feature of the experimental results is that the emitted IR light increases at all pressures with increasing the applied voltage beyond a given threshold, which increases with increasing pressure.

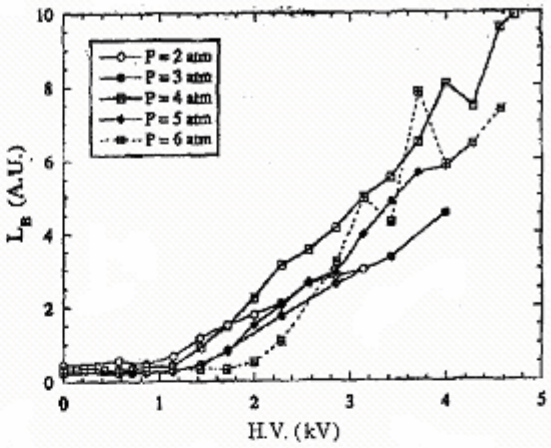

Fig. 8. Infrared light intensity as a function of the applied voltage in Ar gas for several pressures.

For electric fields below threshold the light output is a constant independent of the field. Such a threshold behavior may be explained by assuming that the electrons in the high-energy tail of the energy distribution function are energetic enough to initiate the excitation process.

As a validation test, it is observed that in the respective liquids there is no electric field enhancement of the light output for even larger electric fields because in liquids electrons become epithermal at much higher field strength than in the gas.

Conclusions

We have investigated the beam-induced IR emission by noble gases. We have proved that scintillation occurs also in the IR band.

The most interesting phenomenology of the IR spectra is shown by Xe, where a broad continuum band appears at around 1.3 μm. The position of the centroid of the band and its width depend on the gas pressure.

We ascribed this emission to excimer transitions. A good description of the experimental data is obtained by considering transitions between hydrogen-like levels of the Xe_2 excimer whose energy is reduced by the screening effect of the gas polarization.

Moreover, since the excited states of the dimers are Rydberg states, their energy levels are supplemented by a density dependent con-

tribution arising from the multiple interaction of the outer electron with several gas atoms.

We have some indications that this mechanism is active also in Kr, where measurements are underway.

We have observed scintillation also in the rare gas Ar and Xe liquids and there is evidence that the features of the spectra in liquids are quite different from those in the gas.

Finally, from measurements of IR light emitted during the drift of secondary electrons under the influence of an externally applied electric field, we conclude that the energetic electrons in the distribution may induce similar emission.

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ІНФРАЧЕРВОНЕ ВИПРОМІНЮВАННЯ, ІНДУКОВАНЕ РУХОМ ЕЛЕКТРОНІВ У ІНЕРТНИХ ГАЗАХ ТА РІДИНАХ

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Наводяться перші експериментальні результати з інфрачервоного випромінювання, індукованого проходженням іонізуючих частинок крізь помірно щільні інертні гази. Виявлено багату феноменологію, незалежну від природи падаючих частинок (протонів чи електронів). Виявлено два різні типи поведінки інфрачервоного випромінювання. Вважається, що перший спричинений процесами термалізації вторинних електронів за відсутності прикладеного ззовні електричного поля і пов'язаний зі сцинтиляційними явищами в ультрафіолетовій області. Другий тип – це інфрачервона емісія, що спостерігається при дрейфі термалізованих електронів у газі під дією електричного поля.