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Tunnel ionization of K and L shells model for enhanced X-ray emission by charged up materials

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ABSTRACT

Enhanced X-ray emission from insulator samples charged up during ion beam irradiation, is an effect well known in the particle induced X-ray emission (PIXE) community. Still, its physical grounds remain to be established properly. According to the literature the effect was first observed by Terasawa in 1968, in the early days of PIXE. The effect was to be re-discovered in the 1990s decade, after which a significant attention was put on to it by various authors. Nevertheless, no consistent results and complete generally accepted explanation were reached, so far. Revisiting the problem based on a self-consistent analysis of old data and very recent observations, we have realised that only assuming the occurrence of inner shells tunnel ionization is it possible to explain all experimental details, in particular some that have not been properly justified until now. This conclusion, presents itself of major importance for the PIXE and ion beam communities, but furthermore its relevance extends well beyond this frame by bringing to light, the occurrence of tunnelling ionization of deep energy levels, induced by ion beams, and without the intervention of intense laser beams.

The phenomena of enhanced X-ray emission from insulator samples charged up during ion beam irradiation, was initially assumed to be linked to the discharging process.

According to Kawai et al. [1], the effect was first observed by Terasawa in 1968, who established a set of related patents, in recognition of its importance. The effect was rediscovered by various authors in the 1990s decade [2,3], including one of us [4,5]. After a paper by Peisach et al. in 1993[6], the discussion gained momentum, in particular by Pillay et al. (check [7] and references in it). Soon, it became clear that, whatever the physics, the process (or processes) involved, took place during the charged up stage of the target [2], and not during the discharge.

Observations reported by one of us in a 1998 paper [4], extend its roots to results obtained during the analysis of samples for an intercomparison work aiming at the establishing of a milk powder reference material, still in the end of the 1980s. The esoteric nature of the results delayed the report of the observations until the end of the 1990s.

In both the 1998 [4] and the 2000 reports [5], it was shown, once again, that the effect takes place during the charged up state of the samples.

In the 2000 report [5], an interpretation of the process as due to ionizations by high energy electrons, accelerated within the sample, was presented as most plausible. In spite of this, a cautious warning was made, based on the values for ionization cross-section of keV electrons collisions. It was stated that, ”If we assume that we have equal numbers of protons and electrons, the intensity inversion of the L and K lines …. for the non-neutralised spectrum comes naturally. However, to account for instantaneous enhancement factors we must assume that the number of electrons goes up to 5 to 10 times the number of protons.”.

It was thus clearly assumed that the subject was not closed, even though, at that time, a consensus was installed, in the PIXE community, regarding high energy electrons as the answer to the problem. In fact, if one assumes that some pyroelectric process [8,9] takes place during the irradiation of the sample, in a first analysis of the results, the effect seems to be totally explained.

This generalised consensus, made that in reference [5], some details on the experimental data slipped without being noticed or, at least, properly commented, neither by the authors nor by the reviewers.

Fig. 4 of reference [5], presents a time dependent MSCR record of the simultaneous emission of Zr K X-rays and of bremsstrahlung in the 45 to 100 keV region, collected during an irradiation of a ZrF_4 target, using an 1.0 MeV proton beam, and no electron spray compensation for charging up. A careful analysis of this figure shows that high energy

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electrons can hardly justify all the enhanced emission of Zr K X-rays, because the enhanced emission of Zr K X-rays precedes, significantly, the start of the enhanced high energy bremsstrahlung emission. Furthermore, taking a time interval around 30 s corresponding to a burst of enhanced Zr K X-rays emission, and another around 45 s, it can be seen that while the enhanced emission of Zr K X-rays grows by a factor of about 2 (from the 30 s burst to the 45 s burst), the enhanced emission of high energy bremsstrahlung grows by a factor of 15. Finally it can also be seen that the high energy bremsstrahlung emission intensity peaks towards the end of the enhanced Zr K X-rays emission state, indicating a relation to the breakdown of the process, rather than to its starting up.

The presence of a large number of high energy electrons, generated eventually by a pyroelectric process, therefore cannot be in the origin of all the enhanced X-ray emission, even if plays an important role in the overall process. The fact that the enhanced X-ray emission is more intense in presence of high energy bremsstrahlung enhanced emission, shows that high energy electrons do play a role in the process, still, since high energy bremsstrahlung is not present when the enhanced X-ray emission starts, high energy electrons cannot be the trigger of the process.

Another complex behaviour identified in 1993, is also important to take into account. Fig. 1 shows the emission of X-rays from an FeF_3 sample irradiated by 175 keV protons, without electron spray for charging up compensation. After 120 s of irradiation, when the sample was in an enhanced X-ray emission condition, the irradiation was interrupted by placing a tantalum stopper in the beam path. The X-ray emission did not promptly stopped, but instead presented a strange decay of the emission intensity, having a first fast decaying exponential component and a second more complex component, which can be described by an exponential with an exponentially decaying exponent.

Such a behaviour is compatible with a slow reduction of strength of the process that is promoting the emission, which clearly shows that enhanced X-ray emission is not produced directly by the ion beam.

These old results were very recently complemented by the observation of a significant emission of osmium L X-rays during a test irradiation using an 1.0 MeV 0.5 nA O^{3+} beam at the C^2TN HRHE-PIXE facility [10], in the frame of an IAEA Coordinated Research Programme work [11]. No high energy bremsstrahlung was observed this time, although the system is able to detect X-rays up to 120 keV, and therefore, once again, high energy electrons can hardly be responsible for this X-rays enhanced emission.

The prompt recognition of the importance of this result, made that the first observation was captured in the photo shown in Fig. 2. In this photograph the value of the energy of the L_β X-rays of osmium is shown in the top right conner, corresponding to the position of the cursor. The image of the first author taking the picture is also seen reflected.

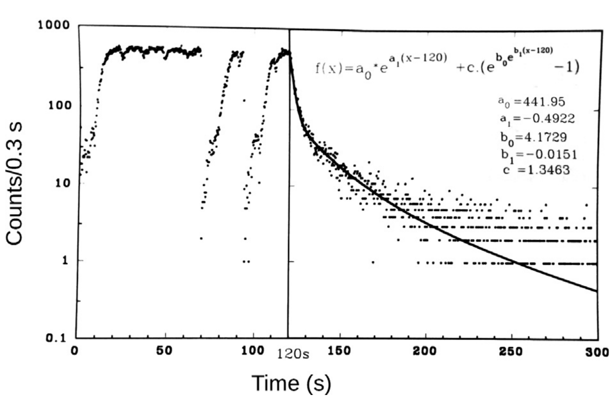

Fig. 1. Multi-Channel Scaling Record (MCSR) spectra of a not neutralized FeF_3 sample irradiated using a 175 keV proton beam. The beam was stopped after 120 s of irradiation, still X-ray emission remained for more than 180 s. Two decay rates are observed, one of about 2 s and one of about 66 s.

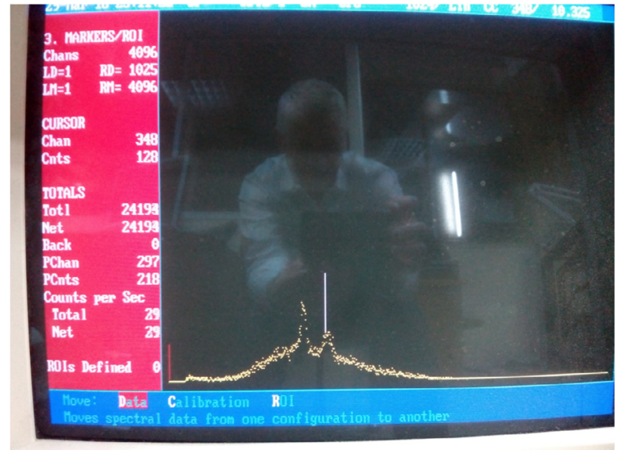

Fig. 2. Photo of the Os L X-rays spectrum obtained during the irradiation of a pellet sample made of pressed 99.995% pure Os powder, irradiated using a nano Ampere beam of O^{3+} ions accelerated to 1 MeV at the CTN 3MV tandetron [20]. The Coulomb ionization of Os L sub-shells by 1 MeV oxygen ions is essentially null. The first author image, taking the picture, is seen reflected on the screen of the monitor.

It was noticed at that moment that an existing electron spray unit used for cancellation of charging up, was loosing intensity. After the replacement of the full capacity of the electron spray unit, the effect vanished and no X-rays could be observed in the same irradiation conditions.

Similar results were obtained by irradiating a pellet of SeO_2. But, it is important to notice that selenium dioxide is an insulator, while osmium is a metal.

This experiment was carried out to check whether Pauli excitation or molecular orbital excitation processes [12,13] could justify observations reported by one of the participants [14] in the mentioned IAEA CRP [11].

In Figs. 3 and 4, spectra obtained in various charging up (Charged) and no charging up (Not Charged) states are presented. The vertical scale was selected to avoid problems related to the accountability of ion beam particles charge.

Since spectra were collected without modifications of the collecting geometry, data from these spectra, allows to estimate values of equivalent cross-sections, from comparison to the results from proton beam irradiations. Values of 24 barn for the Os L_3 shell ionization by 1.0 MeV O^{3+} ions, and 1.5 barn for Se K shell ionization by 1.25 MeV O^{3+} ions, are determined in this way. These values are more than 3 orders of magnitude larger then what can be calculated from data present in Geretschläder et al. [13].

Taking the case of the *Not Charged* 1.25 MeV O^{3+} ion beam, the calculation based on the comparison to proton beam irradiations provides a value of 0.2 barn for Se K shell ionization, implying that even in this case there must have been a residual charging up condition of the sample, as the equivalent value is still much too large.

Highly energetic electrons are not present, since the background at higher energies is nearly zero and the charging up signature in the spectra can only be seen by an enhanced bremsstrahlung present under the characteristic peaks, which extends not much above the energy values of the characteristic transitions.

Verifying the possibility of high energy electrons intervention, it can be estimated from Figs. 3 and 4 that the maximum energy possible for electrons in any of the cases is about 20 keV, which is the end value of the small bremsstrahlung background observed. Taking into account the work of Bote et al. [15] based on PWBA and DWBA calculations for ionization cross sections by electron impact, values of 600, 200, 130 and 110 barn, are determined for osmium L3, L2 and L1 sub-shells and Se K shell, respectively. Now, 20 keV electrons can only produce one