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Intraband luminescence excited in new ways: Low-power x-ray and electron beams

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ABSTRACT

Hot intraband luminescence (IBL) was observed under excitation by high-power (120 keV, 10 A/cm^2) pulsed electron beam, low-energy (10 keV) and low-current (2 μA) continuous electron beam as well as pulsed x-rays. Thus, the fundamental possibility of IBL excitation by x-rays was confirmed, and for most studied materials the absence of the dependence of IBL spectral shape on excitation energy was revealed. For the first time, the intraband luminescence was monitored under low-power excitation, which confirmed the absence of a power threshold of its mechanism and completely ruled out the role of excitation density effects in IBL formation. The data obtained allow the prediction that the IBL can be excited by single photons of 511-keV energy, which is required for enhancing scintillation time resolution in TOF-PET.

1. Introduction

Intraband luminescence (IBL) is a prompt emission originating from the radiative intraband transitions of electrons (e-IBL) and holes (h-IBL) during thermalisation following their creation by ionizing radiation [1]. Despite its relatively low yield the IBL can hopefully enhance scintillation time resolution by providing an almost instant time marker for the scintillation event, for instance, in time-of-flight positron emission tomography (TOF-PET) [2]. TOF-PET requires the detection of single photons of 511-keV energy with exceptionally high time resolution (10–100 ps). IBL is capable of providing such time resolution, as its decay time is determined by the interplay between the probabilities of radiative transitions and phonon relaxation (thermalisation) of charge carriers, which takes place within the picosecond time scale. However to date, there have been no reports on the reliable observation of IBL under 511-keV photon excitation. The radioluminescence decay curves of some scintillators have been studied with sufficient time resolution to detect prompt emissions, however, the IBL could not be distinguished from the Čerenkov light produced by secondary electrons [3]. Therefore, a Čerenkov-free experiment is required to figure out whether IBL can be excited by x-ray or γ-radiation.

So far, the primary excitation sources for studying IBL have been the pulsed electron guns utilizing the field electron emission effect [4]. Scarce complementary data were obtained with a pulsed laser excitation [5]. Pulsed electron guns provide excitation pulses with relatively high peak power. For example, the peak electron current density was up to 2000 A/cm^2 in [4], 100 A/cm^2 in [6], 10–100 A/cm^2 in [7], while our laboratory setup provides about 50 A/cm^2 [8]. Electron pulses of high peak power can be expected to create electronic excitations at high densities, however, the currently accepted model of IBL [1] does not consider it as an energy density effect; neither power threshold for its excitation is expected. Up to now, no observations of IBL have been reported under low-peak-power excitations.

The observations of IBL under x-ray excitation have been scarce and the results controversial. In [9,10], a prompt wide-band cathodoluminescence (CL) of Al_2O_3 has been reported to depend on the energy of exciting electrons. The spectra presented in a wavelength scale were flat in the range 200–1400 nm under excitation by the 300 keV electrons. When the excitation energy was lowered, the CL spectrum narrowed so that at 7 keV the flat spectrum transformed into a narrow band peaking at 380 nm (3.2 eV). The emission spectra of Al_2O_3 under soft x-ray (1–3 keV) and femtosecond VUV laser (16.6–18.2 eV) excitations were also represented by a similar narrow band [11,12]. However, the electron beam currents as well as x-ray and laser power in these works were up to 2000 kA/cm^2 and 10^{12}–10^{13} W/cm^2, respectively, which is significantly higher than in all the works cited above. Other

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authors [6] have reported different results on the spectrum of the prompt ($\tau < 20$ ps) component of the CL of Al_2O_3, which demonstrated two peaks at 2.05 and 2.5 eV. No observations of the spectra of IBL have been performed with the x-ray excitation energies above 10 keV and densities below 10^8 W/cm^2.

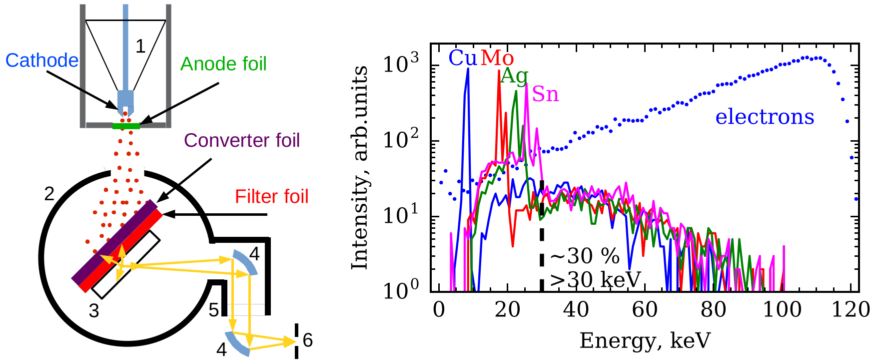
The present research is aimed at the observation of intraband luminescence under low-current electron beam and pulsed x-ray excitation in order to confirm the possibility of IBL excitation with x-rays in principle and to demonstrate the absence of low-pulse-power threshold for IBL emission.

2. Experimental

2.1. Apparatus and setup

The luminescence spectra under high-power pulsed electron beam and x-ray excitation were recorded using a pulsed electron beam setup described in detail in [8]. It was build around a RADAN-303A electron gun, which works by applying pulsed high voltage (100–200 keV) across the cathode-anode gap. During a sub-nanosecond (~300 ps) voltage pulse, the beam is formed due to the field electron emission effect. In this work, we used electrons with maximum energy of 120 keV either directly as an excitation beam or with a converter foil to obtain x-ray pulses (Fig. 1, a). In both cases, the excitation was strongly non-monochromatic. The spectrum of a generated electron beam was estimated using the time profiles of the cathode-anode gap voltage and electron beam current, carefully synchronized taking into account all delays in the measurement circuits. The voltage profile gives the energy of electrons in a particular time moment, while the beam current is proportional to the number of electrons emitted at that moment. The histogram of voltage values distribution weighted by current values is an approximation of the electron spectrum reaching the anode foil. Such approach was used for similar electron guns in [4] and [13]. The spectrum of electron beam in the sample chamber after passing through the anode foil (25 μm Al) was calculated using the Geant4 library with Livermore physics model [14] (Fig. 1, b). In x-ray luminescence experiments, x-rays were obtained by bombarding a 25 μm Cu, Mo, Ag or Sn converter foil with the same electron beam. The filter foil (250 μm Be) was placed directly after the converter foil to absorb any remaining electrons from the beam in order to obtain a pure x-ray excitation. Both the Geant4 simulations and the measurements of the charge accumulated by a Faraday cup placed instead of a sample have shown that the amount of electrons passing the filter is insignificant. Geant4 was also used to model the spectrum of x-rays hitting the sample (Fig. 1, b) for various materials of converter foil. The spectrum is dominated by the characteristic emission line, which constitutes about 40–50% of the total intensity, while the rest is a higher energy braking radiation with a broad spectrum. Unlike the characteristic emission, the braking radiation hardly depends on converter foil material and comprises the photons with energy above 30 keV, which constitute about 30% of the total number of all emitted photons. The peak electron current at the anode foil was 60 A/cm^2, and pulse width ~250 ps. The quantum efficiency of electron to x-ray conversion was estimated by Geant4 as 2×10^{-3}. In cathodoluminescence studies, the variation of electron current (and thus excitation density) was realized by moving a sample mounted in a cryostat further away from the anode foil. In the case of both x-ray and electron beam excitation, the front sample face was irradiated, while the luminescence photons were registered from the opposite one (Fig. 1, a). Several cathodoluminescence measurements were performed in other geometries in order to compare the results of different experiments (see Section 3.2). A reflective collimator was used to collect light and focus it onto the input slit of an Andor Shamrock SR303i spectrograph. This collimator provides a significant improvement in light collection efficiency compared to the quartz lens used in our previous work [8]. It also substantially reduced the radiation load on the cryostat optical window. For light detection, Hamamatsu R3809U-50 MCP-PMT or H10330A-75 NIR detectors were used in pulse current mode with a LeCroy SDA 760Zi-A oscilloscope. An iStar DH 720_18 mm gated iCCD camera was also available for the study of time-resolved spectra. The further details on the detector operation and data treatment have been given in [8].

The CL spectra under continuous and pulsed low-current excitation were obtained using a continuous monochromatic electron gun (Kimball Physics EGG-3101, $E_e = 10$ keV, $I_e = 2.0$ μA) with a focused beam (~1 mm^2 spot on the sample). An ARC Spectra Pro 2300i monochromator with a Hamamatsu H8259SEL photon counting head (dark count rate <3 cps) were used as a registration system in the UV-visible range. For the study of emission decay kinetics the gun is equipped with a beam blanker, which forms 10 ns wide square pulses with the repetition rate of 5 kHz. The peak electron current in the pulse does not exceed 2.0 μA. A Becker& Hickl MSA-300 Multiscaler card was used for decay curve recording. All decay curves were recorded with an equal exposure of 10^6 pulses. A 3 nm platinum coating was applied to the sample surface, preventing the accumulation of electric charge during the measurements. The continuous glow of the heated Y_2O_3 cathode of the electron gun was recorded separately and subtracted from the measured emission spectra. Its impact is low and noticeable only below 2.5 eV (Fig. 5, curve 5). In this setup, the electron beam was exciting the same sample face luminescence photons were registered from (see Fig. 7, geometry 1).



(a) (b)

Fig. 1. The experiment layout for x-ray luminescence (a), and the Geant4-simulated spectra of various excitation beams (b). The anode foil is 25 μm Al, the converter foil is 25 μm Cu, Mo, Ag or Sn, and the filter foil is 250 μm Be. For cathodoluminescence experiments the converter and filter foils are removed. 1-electron gun, 2-vacuum cryostat, 3-sample, 4-off-axis parabolic mirrors (Al+MgF_2), 5-fused silica window, 6-monochromator input slit.