Radiation-induced inelastic processes in Rare-gas Solids probed by VUV luminescence spectroscopy

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The modification of material properties via electronic excitation and relaxation processes is an important prerequisite for many novel technological applications in material science, microelectronics, photochemistry and biology. These effects are based on strong interaction between electronic and nuclear (atomic) subsystems followed by a concentration of the electronic excitation energy within a volume about that of a unit cell and the energy conversion into the kinetic energy of atoms in solids. Defect formation induced by electronic transitions (DFIET) is a topic of active current research spread over a variety of materials [1,2]. However, the available up to recently models of this phenomenon permitted only a general qualitative description. Details of these processes including the sequence of elementary stages, their interconnection and competition were not quite understood. This situation was caused by a lack of data on state-selective DFIET.

The main aim of the study was to elucidate primary electronic processes and interconnected relaxation channels resulting in lattice rearrangement through energy concentration in a critical degree of freedom. Radiation-induced atomic processes were investigated in model wide band gap insulators: Rare Gas Solids (RGS). Small binding energies in conjunction with a strong exciton-phonon interaction result in high quantum yield of DFIET that makes RGS especially suitable for the experimental study. This report summarizes the study of permanent defect formation in the lattice via excitonic excitations.

For the state-selective study of DFIET we have used a set of spectroscopy methods because, as obvious, a spectroscopy is the most adequate method aimed to study of the processes induced by electronic transitions. Using Synchrotron Radiation (SR) as a selective excitation source is the most promising way to clear up primary stages, elementary processes and DFIET mechanisms in detail. The experiments were performed at the unique SUPERLUMI station (HASYLAB at DESY). The samples were grown in a special closed cell mounted on a He cryostat holder in an ultrahigh vacuum environment (10^{-10} mbr). High purity gas was condensed at elevated T under isobaric conditions. The initial T of deposition was fixed at about $0.75T_t$ (T_t is the triple point temperature) for each gas, whereupon the sample was cooled down to 5K with a constant rate of 0.1 Ks^{-1}. On the sample preparation the cell was removed and the high quality transparent free-standing RGS were subjected then to a long-term exposure of the VUV photons of chosen energy. The study was focused on the exciton-induced processes of lattice rearrangement.

The novel spectroscopic method of a lattice rearrangement study was developed taken advantages of Synchrotron Radiation techniques. The study was concentrated on processes of defect formation via two channels of exciton self-trapping: (i) self-trapping into atomic type state A-STE and (ii) self-trapping of excitons into molecular type states M-STE. The channel (i) dominates in “light” Ne solids, whereas the channel (ii) is the main channel of the exciton self-trapping in “heavy” RGS (Xe, Kr, Ar). For solidified “heavy” rare gases the well-known M-bands were analyzied. In the case of “light” Ne solids atomic A-bands yielded information on the processes of lattice rearrangement

induced by electronic excitation. Photoluminescence spectra of all classical RGS are shown in Fig.1.

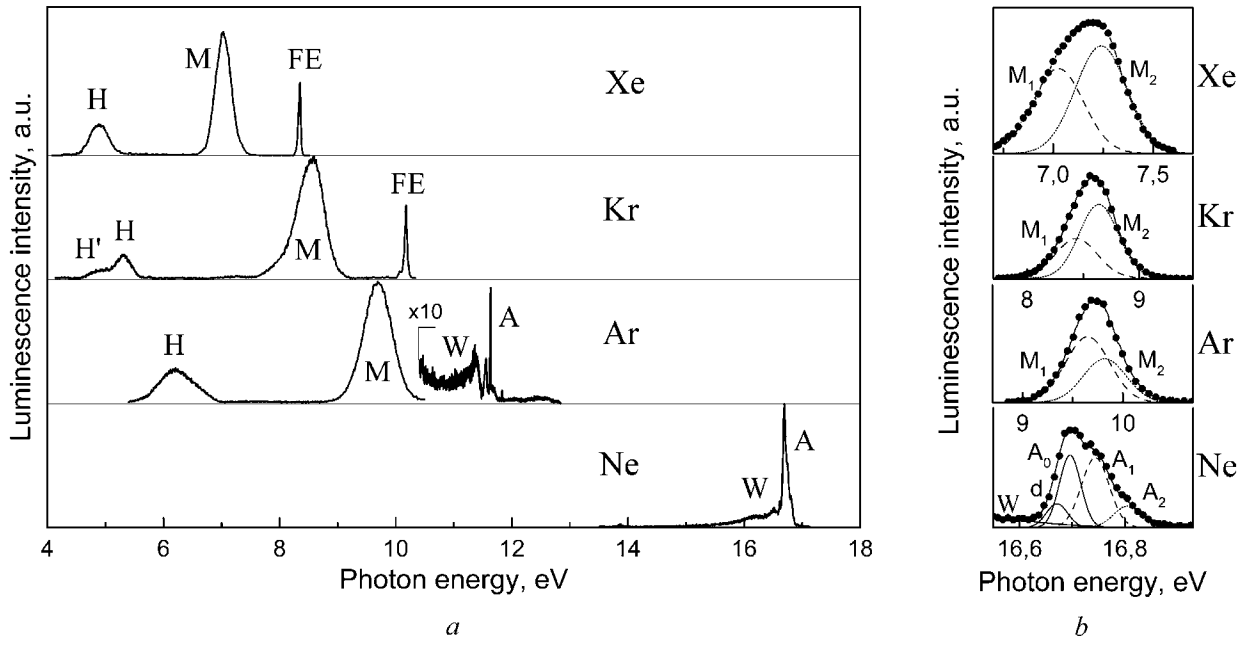

Figure 1: Luminescence spectra of RGS at T=5K; (a) general view of the spectra under excitation by photons in the range of band-to-band transitions; (b) internal structure of M- and A-bands under excitation in the corresponding excitonic range.

As it was found in our previous studies [3,4] the luminescence spectra of both M-STE and A-STE states have their internal structure and consist of defect-related subbands (M_1, A_1) and ones stemmed from the exciton self-trapping in the regular lattice (M_2, A_2). Real-time monitoring of the defect-related features in the luminescence spectra of RGS upon selective excitation with SR enabled us to get the direct evidence of defect (Frenkel pair) formation via exciton self-trapping into A-STE and M-STE states. Verified mechanisms of the exciton-induced lattice rearrangement are classified into two qualitatively different groups: "ground state" mechanism operating after electronic transition to the ground state and "excited state" mechanisms operating during the lifetime of the excited states. The key role of crowdions in the processes of defect formation is suggested.

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