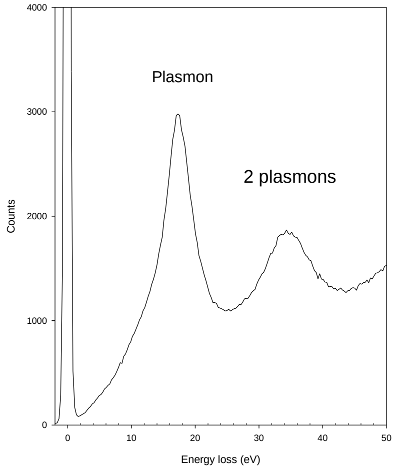
Collective excitation in the ionization of atoms, in semiconductors and biological tissues

Researcher: Dr. Tibor Papp
Report on the work progress and achievements
International reintegration grant FP7-MC-IRG 224938

Except for closed shells, most atomic states exhibit some collective behavior. That is, the states cannot be described as a single product function, but superpositions of product states. In the description of the interaction of radiations with atoms, the independent particle approximation is used. Our angular distribution and angular correlation study, and in synchrotron photoionisation measurements there is a strong deviation from this model. Revisiting previous measurements with an open eye for the possibility of collective excitations, we can explain the majority of discrepancies by it. We have presented all the evidences in a paper and recommended best practices. We also give a critical review calling attention to the problem that theory laden experiments are used to verify theory. We have made on overview of the problems in determining the L shell fluorescence yields and Coster-Kronig transition probabilities. We have made a critical analysis of the experimental methods which are used to determine Coster-Kronig and fluorescence yield data. We have concluded that each of them has unrecognised systematic errors. We have recommended new approaches.

The collective excitation in Si and Ge semiconductor detectors is plasmons. A REELS (Recoil Electron Energy Loss spectroscopy) spectrum for 4 keV electrons is presented here in the inlet of this paragraph.

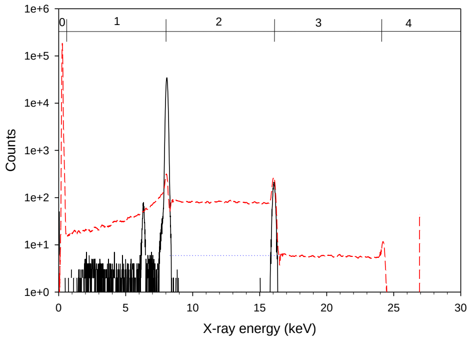


When x-ray or gamma hit a detector, the main interaction is to create energetic electrons. Although, the energy loss of energetic electrons is dominated by plasmon creation, the detector models do not include the plasmon decays. The electron energy loss was studied by REELS (Recoil Electron Energy Loss spectroscopy) with a high energy range electron spectrometer in detector materials InSb, GaAsAl, diamond, in addition to Si and Ge. We have developed a new statistical model for the resolution, when the process has two steps, first a plasmon is created, then the plasmon decays, and derived the Fano factor model for Si and Ge semiconductor detectors.

This is a new approach. We can see from the derived equation that the resolution depends on the plasmon properties. Previously only the band gap was used to search for good resolution detector materials. Therefore the search for detector materials can be revisited, searching for detector material candidates, via screening the plasmon parameters. The detector resolution previously has not been derived including the dominating decay channel, the plasmon. On this base InSb is not a potential candidate for good resolution detector.

In the REELS study on energy loss processes in Si and Ge detector crystals, we have observed the plasmon dispersion, and from the plasmon dispersion the difficulties in semiconductor x-ray detector energy and width calibration can be explained. This is a long overdue problem in x-ray spectroscopy with energy dispersive detectors.

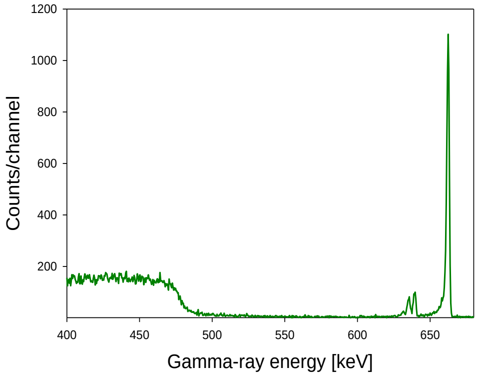
Using a quality assurance digital signal processor we have developed a method to determine the true input rate for x-ray semiconductor detectors. The signal processor inspects the signal coming from the preamplifier and decides that it will be a desired quality signal or an undesired quality signal and reject it. The rejected signal usually counted and indicated in the dead time, and pile up counter. However, the rejected events will include noise events, noise piled up with noise and real events, single-event rejection peaks for those individual events that do not pass the event discrimination tests, multiple-event pileups as well as non x-ray (particle or high energy) events that generate a signal in the spectrum. We have developed a procedure to account for it. It is based on the signal processor capability to analyze all the events and place it to separate spectra. From these the true input rate can be determined. Such a spectrum pair is shown in the figure of this paragraph for a quasi-monochromatic Cu Kα1 radiation detected with a Si(Li) detector. It also explains many new features in the response function, and possible origins of the varying electronic efficiency at several other systems.



We were invited to participate and contribute to the IAEA workshop on future directions in digital signal processing in gamma ray spectroscopy. We have made comparative measurements with the standard system and the CSX quality assurance capable digital signal processor at a gamma ray metrology institute, and in a nuclear research center. Strong differences sometimes a factor of fifteen in intensity was found for the favor of CSX processor. Observation on the origin of background, and change in the Compton shape were found as well. The presented examples demonstrated that even in expert laboratories the detector system adjustment and tuning needs special expertise, and provide clear demonstrations of several of the potential problems facing the analyst. The geometrical and noise environment set up requires experience. The availability of all the events also provides a wealth of information on detector physics, allowing identification of the signals resulting from slow rise time, or distorted signals originating from imperfections. It can readily monitor ones efforts at hunting for sources of system noise. Two additional points; seeing all of the events is very useful in education and training of future analysts; as well as seeing the separation of events between accepted and rejected spectra can aid in the design of experiments, e.g. a Compton plateau may indicate the improper positioning of a shielding material.

We have designed and built a circuit to transform the decaying type preamplifier signal to staircase like signal, which is more favorable for digital signal processing.

With a quality assurance capable digital signal processor we have obtained excellent quality spectra for gamma rays using a CZT (cadmium zinc telluride) detector. A spectrum of the 661.65 keV gamma line of ^{137}Cs, measured with a CdTe (3x3x1 mm^3) is presented in the left side of this paragraph. The well-resolved escape peaks and

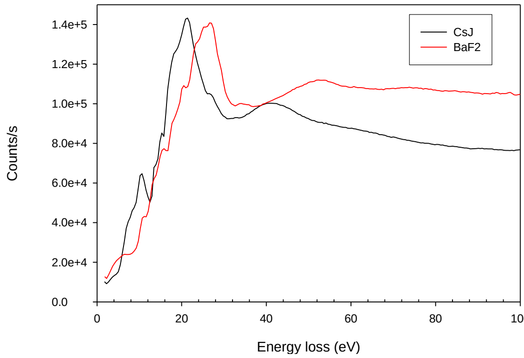


the small low energy tailing demonstrate the resolution capability. It is very promising; keeping in mind that hole transport is a major limitation on device operation, as with many other compound semiconductor materials.

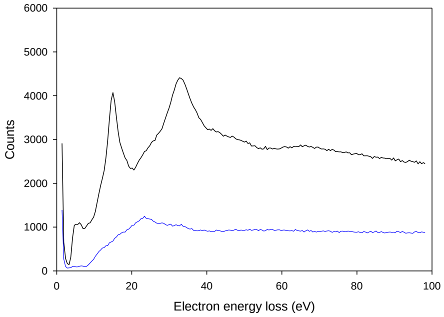
REELS measurements on CdTe detector materials revealed that the dominating energy transfer quanta are related to collective excitations, and the CdTe material could give very high quality spectra. Comparing the REELS spectra of CZT with Cd and Te it seems that three different type of collective excitation is in present of the CZT detector material.

We have measured the energy loss processes in scintillator crystals. We have discovered that the basic energy loss process is giant resonances. We have found giant resonances in LSO, LYSO and GSO and several other scintillators, which are frequently used in medical imaging, including positron emission tomography (PET). The giant resonance allows the interpretation of several properties of the scintillators, like temperature dependence, oxygenation, photon energy, radiation damage. As the giant resonance is very sensitive to the potential shape, the strong sensitivity on the atomic environmental variation can be explained. Turning the argument around, the scintillator properties can be designed by tailoring the potential supporting the giant resonance. We have made heat treatment on scintillators, and we could see the change in the scintillator by the heat treatment.

We have extended the study of dominating energy transfer modes in scintillation crystals, of CsI and CsCl. In each case we have found that there is a collective excitation which is the dominating energy transfer. When we have divided the gamma ray energy with this excitation energy we obtained the photon number for CsI, indicating that one photon is created from one excitation. This is a nice outcome. The REELS spectra are presented for BaF2 and CsI in the figure at the left. Similar spectra for CsCl indicates, that CsCl has the sharpest collective excitation peaks, and the longer term inelastic contribution is smallest, therefore perhaps this material could have very novel properties as a scintillator.

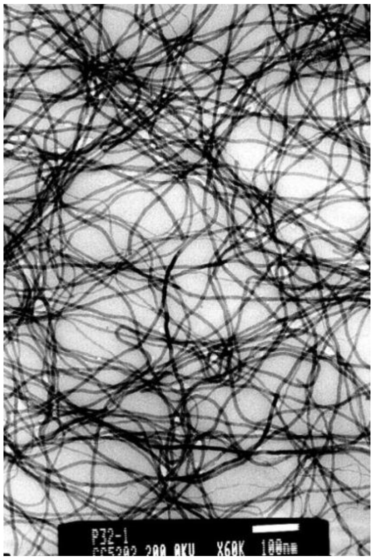


Observing that the collective excitation is the dominating energy loss in semiconductor detectors and scintillators, and in some of the cases the giant resonance excitation is dominating, we searched for giant resonance excitation in rare earth compounds and observed it in each case. For potential biomedical use, we have searched for those compounds which are used in homeopathic medicine, with high dose tolerance. This turned out to be a rewarding idea, as the strongest giant resonance excitation was found among them. An example is presented in the figure beside, demonstrating how much a giant resonance can change in different chemical compounds. We have concluded, that all the studied detector materials having good resolution can be described as the electron energy loss process is dominated by an energy path, which is mediated by a collective excitation, and the energy and breath of the

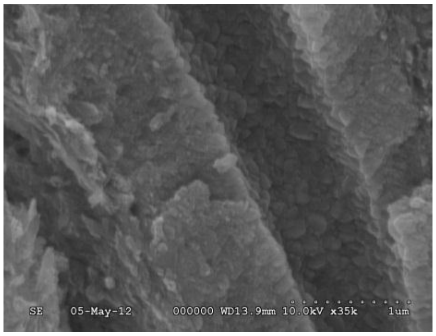


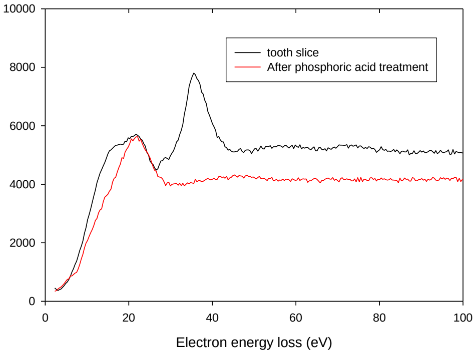
collective excitation is a factor determining the resolution. Therefore it is desirable to tailor the collective excitation.

It is most promising to tailor it with using nanomaterials, where the size of the nanomaterial will modify the properties of the giant resonance. Therefore we have experimented with nanomaterials and manufactured tellurium nanowires. We have studied the giant resonances in telluride compounds, and made telluride nanostrings. Large variation of the giant resonances was observed for the different size and shape of the giant resonance. We were successful to grow telluride nanostrings with large aspect ratio, 6 nm diameter and 40 mm in length. The transmission electron microscope image of tellurium nanostings is at the right side of this paragraph.



We have made XPS and REELS spectroscopy of permanent adult and children's deciduous teeth. Collective excitation of Ca compounds were found. Large set of pictures were taken by an electron microscope, a picture of nano-aggregates in the dentinal tubule is seen at the left. There are nano-aggregate components in teeth. Using the standard phosphoric acid treatment as dentist use, the giant resonance disappeared. At the left such REELS spectra are presented before (black line) and after treatment (red line). The red line was scaled up to show a shape overlap. The difference corresponds to nonorganic components. We have found giant resonances in modern the tooth fillings used in dentistry.





Deriving the new statistical model for the resolution limit can trigger the search for new detector materials with novel properties on a new principle. Understanding the detector can yield better designed measurements and more accurate data. Quality assurance capable signal processing increase the capability and reliability of basic research and analytical works in x-ray and gamma ray techniques, which is broadly used in industrial processing, mining, environmental, and medical field as well. This is also necessary to assure that the methodology is capable to be used in verification of compliance with the legislated requirements.

We have found strong presence of collective excitations and giant resonances in a broad variety of fields. It seems that it is a very general rule in nature, when a collective excitation is available the energy transfer will be mediated by the collective excitation channel. Giant resonances can effectively channel the energy loss path; therefore it can be used to specifically design the energy loss in biological matter as well.