NOMATEN Hybrid Seminar

Location: PNT room 223 (Neutron) Time: 1 PM gotomeeting room (for online): <u>https://meet.goto.com/NCBJmeetings/nomaten-seminar</u> Seminar date: November 2nd, 2023

Title: Alpha-decay damage in natural apatite: buildup and recovery mechanisms for thermochronological applications

Speaker name: Dr. Frederico Garrido

Speaker affiliation: Université Paris-Saclay, IJCLab - Laboratoire de Physique des 2 Infinis Irène Joliot-Curie, Accelerator Valley, Orsay Campus, France

Abstract: Thermochronology is an earth science discipline involved in the reconstruction and timing of past geological processes. In this field, rock accessory minerals such as apatite are collected and their thermal histories (temperature vs. time) reconstructed to study landscape evolution and mountain-building processes. One of the low-temperature techniques useful in the thermal history modeling of rocks is apatite (U-Th)/He thermochronology. Under this technique, an apparent thermal age termed as "He age" is measured for each apatite crystal via analytical measurement of its present-day U, Th, Sm, and He contents. For each crystal, the measured amount of He inside is calculated as the difference between He production and diffusional losses: (1) He production from the alpha decay events of Sm and those of the natural U and Th radioactive decay series less the (2) amount of He that diffused out of the crystal at high temperatures.

The diffusion coefficient of He in apatite, which affects the measured He age, is based on the Arrhenius Law. Recent studies, however, strongly indicate that the amount of radiation damage in a crystal modifies the He diffusion coefficient, and consequently, the measured amount of retained He used in the thermal history modeling of rocks. The quantification of the amount of radiation damage, which is primarily due to alpha decay, and the modeling of kinetics and mechanism of damage buildup and recovery in apatite, are thus of great importance in the modeling of thermal histories. The aim of this research is to investigate in a fundamental level using in situ and ex situ ion implantation, Rutherford Backscattering Spectrometry in channeling mode (RBS/C) and Monte Carlo simulations using the McChasy code, and Transmission Electron Microscopy (TEM), the kinetics and mechanism of the following physical processes linked with alpha decay: (1) radiation damage buildup separatelyinduced in apatite by the recoil nuclei and alpha particles, and (2) the athermal, ionization-induced recovery of pre-existing defects attributed to the electronic energy loss of alpha particles. The damage buildup experiments in the atomic (nuclear) regime were conducted over a wide ion fluence range to fully characterize and model the irradiation effects of alpha decay in natural apatite minerals. Both 30 keV He and 500 keV Bi ion implantation on (0001)-oriented natural Ca₅(PO₄)₃F single crystals were used to mimic the radiation damage at various stages. The irradiation temperatures utilized in the experiment were also varied to correspond with typical thermochronological applications (in the range 293-393 K). Concerning the ionization-induced recovery caused by alpha particles, experiments were extensively conducted with the objective of determining the dependence of damage recovery kinetics with the initial amount and type of pre-existing defects in an apatite crystal. For this purpose, 1.4 MeV ions were used to both collect RBS/C spectra to measure the defect concentration using Monte Carlo simulations but also to induce ionizing-induced recovery of defects generated by atomic collisions.

The RBS/C Monte Carlo simulation results and TEM characterization of this study strongly suggest that the buildup of radiation damage in apatite as induced by both (1) alpha recoil nuclei and (2) alpha particles could be described as a two-step kinetic process, and that each step could be characterized by the presence of specific main types of irradiation-induced defects. The first step is a transformation from a near perfect crystalline structure to a partially damaged one, while the second step, which is triggered after a certain damage dose, results in a further structural transformation leading to a highlydamaged state. Amorphization due to alpha recoil irradiation and helium bubble formation due to alpha particle irradiation are the two physical mechanisms that led to rapid apatite crystal destabilization. With regard to ionization recovery, experimental results suggest that only certain types of defects such as amorphous clusters could be efficiently recovered by alpha particles and that point defects are likely less sensitive to the recovery effect. This means that alpha-particle induced recovery of pre-existing defects does not occur or is likely insignificant for natural apatites with very low U and Th concentrations (few hundreds of ppm). Owing to the significant implications of this irradiation study to the methods utilized by the thermochronological community, the obtained kinetic models will be incorporated in a later stage in common thermal history modeling codes to perform the next step of geological calibration and applications.

Bio: Prof. Frederico Garrido's research focuses on fundamentals of ion-solid interactions and radiation-induced damage in materials, behaviour of ceramic-type materials under irradiation with a special emphasis put on nuclear materials (fuels, transmutation matrices and specific immobilization matrices for high level nuclear wastes) and their performance in extreme environment, mechanisms of phase transformation under irradiation, understanding of defect evolution at atomic scale, synergetic effects in dual beam irradiation. He is currently Associate Scientific Director of the Pole Energy & Environment at IJCLab located at the Orsay campus.