Crystal assisted experiments for multi-disciplinary physics with heavy ion beams at GANIL

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Abstract. We present a review of the channeling and blocking experiments that have been performed at GANIL during the 30 years of stable beam operation, with the strong support of the multi-disciplinary CIRIL-CIMAP laboratory. These experiments combine atomic physics, solid-state physics, and nuclear physics.

1. Introduction: Channeling and blocking of swift heavy ions
Briefly, ions travelling in a crystal with a propagation direction close to a major axial or planar direction undergo a series of correlated collisions with aligned atoms, which leads to a non-uniform flux and anisotropies in the angular distribution of the particles inside the crystal [1, 2]. At sufficiently large distances from atomic strings or planes, i.e. in conditions where no nuclear or large angle elastic collisions occur with target nuclei, the continuum potential approximation allows one to describe channeled ions as particles trapped into a 1D or 2D potential well, in planar or axial channeling conditions, respectively. Particles at the crystal entrance are given a transverse energy $E_t$ that depends on the incidence angle $\psi_{in}$ and the transverse position $r_{in}$:

$$E_t = E \psi_{in}^2 + qU(r_{in}).$$

If $E_t$ is small enough, typically $E_t < qU(\rho_{th})$ where $\rho_{th}$ is the thermal vibration amplitude of the crystal atoms, then one can consider that ions are channeled, and keep their transverse energy throughout the crystal in first approximation. The above condition was expressed in terms of a critical angle by Lindhard [3]. In the case of axial channeling, the incidence angle of the beam relative to the axis should be smaller than $\psi_{ci} = (2Z_1Z_2e^2/Ed)^{1/2}$.

The blocking effect corresponds to a reduction of the probability, for particles emitted close to atomic sites of the crystal, to emerge in a direction parallel to crystallographic ones. Stricto sensu, channeling and blocking are reciprocal effects, and they are governed by the reversibility rule: both correspond to a redistribution of the ion flux inside the crystal. Channeling leads to a reduction of close collisions with atomic cores when a parallel beam is aligned along a crystallographic direction, whereas blocking is a reduction of the flux parallel to a crystallographic direction for ions emitted at close distance from an atomic core. The same characteristic critical angle governs both processes. A particular application of blocking is the measurement of nuclear lifetimes at the attosecond scale by measuring transverse nuclear recoils with respect to atomic strings or planes.

2. Material and method
The GANIL accelerator is a unique facility to provide high intensity, low emittance beams of heavy ions at a few tens of MeV per nucleon. Ions produced in an ECR source are injected in a first cyclotron C0, and then in a first separated-segment cyclotron CSS1, after which they reach 5-6 MeV/u for the heaviest species, then stripped in a thin foil before injection into the second CSS2 cyclotron, reaching 24 MeV/u for U beams. High energy ions emerging from CSS2 have typically the most
probable charge state of the medium energy after the stripper foil, i.e. they are hardly fully stripped for $Z>20$. Typical intensities of 10-100 nA are available within an emittance value of $15\pi \text{mm}\times\text{mrad}$. The maximum intensity depends on the nature of the ion species (rare gas ions are produced much more easily than metallic ions for instance). Further stripping and energy degrading may be performed by inserting a target of suitable thickness and composition at the exit of CSS2. In order to realize detailed channeling experiments, strong reduction of the emittance has to be performed. This is achieved upstream and downstream of the GANIL’s alpha spectrometer, by means of two series of three slits and dedicated quadrupoles. The momentum dispersion of the beam may be reduced at the dispersive plane of the alpha spectrometer by means of another slit. The whole alpha spectrometer is achromatic. After acceleration and tailoring of the beam with adequate emittance (of the order of 0.1 $\pi \text{mm}\times\text{mrad}$ in both directions), energy spread, charge state and intensity, the ion beam is guided and distributed into the various experimental areas.

The first experiments took place in the LISE beam line, for which beam optics was optimized for channeling. It comprises a dipole magnet with a dispersion of 1.8 cm/%dp/p at the focal plane, and a vertical-slit collimation to perform charge- or energy-selection downstream of this dipole. A multiwire gas proportional chamber (MWPC) could be used for transmitted beam profile acquisition, and therefore to energy loss and charge state distribution measurements. Later, the high-resolution SPEG was used (8 cm/%dp/p at the image plane) equipped with position sensitive MWPC with event-by-event detection capability.

3. Experimental program

One of the particularities of the ions accelerated at GANIL is that heavy ions are not fully stripped, and charge exchange plays an important role in particle matter-interaction. Another is that high electronic energy loss regimes are achieved with swift heavy ions. This motivated the implementation of atomic and solid-state physics research programs, hosted by the CIRIL laboratory.

As soon as GANIL was built, a large manifestation of interest took place among several multidisciplinary laboratories for setting up channeling experiments. These experiments were hosted by the CIRIL (now CIMAP). The initial collaboration gathered solid state physicists from the Groupe de Physique du Solide (Paris, later renamed as INSP), Laboratoire des Solides Irradiés (Fontenay aux Roses), atomic and nuclear physicists from Institut de Physique Nucléaire de Lyon, Centre de Recherche Nucléaire (Strasbourg), and a bit later Centre d’Etudes Nucléaires de Bordeaux. Beam experts from GANIL were also contributing. The IPNL, INSP and CIRIL groups have been continuously working on this program during more than 20 years.

The first GANIL-channeling publication, by Camille Cohen et al. [4] dealt with 60 MeV/u Ar$^{18+}$ ions channeled in a Ge crystal. Products of nuclear reactions (gamma, neutrons) were used to characterize the extinction of close collisions, and the observed values of critical angles and extinction yields were found – as expected – in agreement with extrapolated values from lower energy experiments.

3.1. Charge exchange, energy loss and related surface-emission studies

In amorphous solids or random crystal orientation condition, Nuclear Impact Ionization (NII) and non-radiative electron capture, also called Mechanical Electron Capture (MEC), are the dominant processes in the charge exchange by heavy ions in the high-energy domain covered by GANIL. In contrast, charge exchange by channeled ions is dominated by ion-electron interaction processes that do not involve nuclear recoil during close collisions. In that way, the crystal target can be considered as a dense and non-homogeneous electron target. The various processes that have been studied at GANIL are presented in figure 1, with a representation of the electronic transitions in the ion energy frame.
The first charge exchange study dealt with Radiative Electron Capture (REC) by hydrogen-like xenon ions at 25 MeV/u, which lead to a measurement of K- and L-REC cross sections [5]. As the adiabaticity parameter \( \eta_K = \left( \frac{v_p}{Z_p v_0} \right)^2 \) (where \( Z_p \) is the ion atomic number, \( v_p \) the ion velocity, \( v_0 \) the Bohr velocity) is much smaller than 1, this means that the K-shell is filled at charge state equilibrium. Thus K-REC cannot be observed in random crystal orientation, because non-radiative electron capture, also called Mechanical Electron Capture (MEC), followed by radiative decay, rapidly fills the K-shell vacancy. Therefore one has to use the property of the extinction of close-collision induced-charge exchange to observe REC, which results from pure ion-electron interaction. This was a first step toward electron capture into inner-shell at out-of-equilibrium conditions. At the same experiment, several incident charge states were used, and the incident charge state freezing in axial condition could be studied in detail [6].

A detailed study of energy loss and Electron Impact Ionization (EII) was reported by A. L’Hoir et al. [7]. In this experiment, 27 MeV/u Xe\(^{35+}\) ions were used as projectile into a 21 µm thick silicon crystal aligned along the <110> axial direction. In this case, the M-shell ionization probability is very high, even for EII, and thus EII rates could be used as a probe for the local electron density evaluation along the ion path in the crystal. Combining these studies with energy loss measurements led to a detailed study of local and non-local parts of the energy loss as a function of transverse energy. One of the conclusions was that, at such not-so-high velocities, the energy loss of low –transverse energy ions is due only to the valence electron gas, however it is almost independent on the local electron density. The extinction of the contribution of silicon L-electrons to energy loss was observed to be more dependent on the local electron density.

In collaboration with the IKF-Frankfurt, electron emission spectroscopy at the crystal exit was performed in 1990, using 27 MeV/u Xe\(^{37+}\) incident ions on a 21 µm thick crystal. The results were only partly published in [8]. An extinction of the binary encounter electron yield (at velocity close to 2 \( v_p \) at 0°) in axial crystal orientation was found equal to the ratio of valence to total electron density in silicon, showing that this yield depends on the local electron density.

**Figure 1:** Charge exchange resulting from ion-electron interaction that have been studied in channeling conditions.
The same year, an experiment dedicated to the dielectronic capture, also called Resonant Transfer and Excitation- RTE in the case of bound target electrons - took place in the LISE beam line. Thanks to a particular effort from the accelerator team, a full resonance was described, with energy variation of a Xe$^{52+}$ ion beam from 42.3 to 33.8 MeV/u, while keeping constant the beam characteristics at the target point. The resonance was observed by means of $K_\alpha$ radiative decay and by charge state distributions as a function of energy loss measured after charge and energy analysis. The shape of the resonance was compared to calculated Compton profiles [9]. This was the last of four experiments performed with the same 21µm thick silicon crystal. Radiation damage is quite reduced in channeling conditions.

The following experiment – during which the atomic physics group of the GPS-Paris joined the collaboration - was again devoted to REC, but this time in conditions where REC would be observable both in channeling and random conditions, i.e. at $\eta$ value greater than 1. This was achieved with krypton ions at 60 MeV/u into a 37 µm thick silicon crystal. Coincidences between X-rays and transmitted ions with given charge state were used to sign electron capture or ionization. Using high statistics and high resolution X-ray spectra, the shape of the K- and L-REC lines were used to infer impact parameter dependent Compton profiles from the various electronic levels of silicon (valence and core electrons) [10]. A by-product of this experiment was the measurement of intrashell excitation which depopulates metastable n=2 states of H-like or He-like ions, even in channeling conditions [11].

Then followed an attempt to observe Trielectronic Capture or RT2E (single electron capture accompanied with double excitation of the ion), namely KK-LLL, by means of triple coincidence between a single capture event and two $K_\alpha$ photon decay. The first attempt, using Nb$^{59+}$ ions in LISE, was interrupted by a fire in the injection cyclotron. The final attempt was the first channeling experiment performed in the high-resolution spectrometer SPEG, using He-like Kr ions, with GANIL researchers. An upper limit for the cross section of this effect was obtained [12].

The next experiment in SPEG took place in 2000. The objective was twofold: to study the dependence of electron emission from both entrance and exit surfaces with the transverse energy of incident Pb$^{64+}$ ions at 29 MeV/u, and make detailed study of energy loss as a function of charge exchange in a very thin crystal (1.1 µm thickness). Backward electron yields were found to be correlated to the impact parameter at the crystal entrance, whereas forward yields were found less correlated to the transverse energy [13]. Another result from this experiment was the observation of the superdensity effect, which leads to ions with higher charge state and higher energy loss than obtainable in random conditions, due to series of close collisions at small impact parameter that enhance the probability of multi-step ionization, and prevent decay into deeply bound states after capture. The corresponding excess in energy loss during close collisions is not compensated by lower electron density sampled at large distance from the strings inside such a thin crystal [14].

In collaboration with CEA-DAM and CENBG-Bordeaux, an experiment was set in the LISE beam line to observe NEEC (the time-reverse process of internal conversion) by $^{57}$Fe$^{26+}$. The aim was to excite the 14.4 keV nuclear transition by resonant capture into the K-shell at 9.4 MeV/u, and to detect the signature of the in-flight decay of the excited level by internal conversion, in the 6 meter straight section between the two dipoles of LISE. The vacuum conditions in the beam line prevented us to reach the sensitivity required to observe the microbarn cross-section-NEEC. So far this process has still never been evidenced, and channeling remains a competitive technique to observe it, since an aligned crystal of a few microns is equivalent to a pure electron target of $\sim$10$^{20}$ e/cm$^2$.

The last experiment performed by the collaboration in 2006 (with IPN-Orsay collaborators) consisted in the measurement of ionic sputtering by the surface of a germanium crystal. The energy loss of an incident ion at the entrance of an axially aligned crystal varies over one order of magnitude as a function of transverse energy. This is expected to influence strongly the ion emission yield. An
ionic mass spectrometer was set at the entrance face of a thin germanium crystal in SPEG, and secondary ions were detected as a function of the charge state of Pb incident ions at 29 and 5.6 MeV/u. With the actual experimental conditions (secondary vacuum), mainly organic molecular ions from the surface were detected. However, we could observe that emission from amorphous compounds covering the surface is strongly correlated to the energy loss below the surface, where channeling acts for differentiating this particular feature of energy deposition [15].

3.2. Resonant Coherent Excitation
Resonant Coherent Excitation (RCE) makes use of the lattice periodicity to induce internal excitation of the projectile. In a collaboration between IPNL-Lyon, GPS-Paris, CIRIL-Caen and CENBG-Bordeaux, a feasibility study to observe nuclear RCE of a low-energy transition of a metastable state of $^{45}$Ti (3.45 keV) was undertaken in 1994. However, since the candidate nucleus has to be produced by means of fragmentation, the beam intensities achievable at GANIL at that time could not allow one to observe this resonance [16].

About the same period, we certainly missed the opportunity to collaborate with Sheldon Datz, who proposed to the members of the collaboration to use planar channeling for varying the excitation frequency during atomic RCE experiments. Using this method, and with the very low momentum dispersion and low emittance achievable with the GANIL beams, precision experiments would certainly have been successful. This way of studying atomic RCE is still employed by the Japanese group of Azuma et al. [17].

3.3. Nuclear fission times
Nuclear physics applications of ion-crystal interaction have been studied experimentally at GANIL. In addition to the search for NEEC and nuclear RCE already discussed above, nuclear lifetime measurements by the blocking technique were performed. The first experiment was done by J. Gomez Del Campo et al. [18] who measured long-lived projectile-like fragments during the bombardment of a germanium crystal by argon ions.

Later, a collaboration between GANIL, IPN-Lyon, INSP-PARIS, CEA-Saclay, IPN-Orsay and CIRIL-Caen performed a series of experiments to measure fission times of uranium and lead during collisions with a silicon crystal at high energy. The use of inverse kinematics allows one to detect both fission fragments. The excitation energy was measured by the ORION neutron detector. In the case of uranium fission, strong variation of the average fission time was found as a function of excitation energy [19]. As the effect corresponds mostly to a uniform filling of blocking dips as the excitation energy decreases, the interpretation by means of simulations relies on an increase of a fraction of long fission times for which no blocking is observed (i.e. when recoil nuclei travel typically more than half of the transverse cell before fission) [1]. For $^{208}$Pb ions that are much less fissile, a trend was found for the existence of non-short lifetimes ($t > 10^{-18}$ s typically) when the excitation energy was the smallest and for fissioning nuclei $Z_1 + Z_2 \leq 82$ (sum of the two fragment atomic numbers) [20]. Note that, in this experiment, the comparative analysis of fission times and elastic scattering recoils (used as reference for instantaneous events) was made for blocking along major planes, one of which being almost coplanar with the beam direction. Therefore, the time needed for a nucleus to recoil at distances larger than the thermal vibrations is increased by one order of magnitude with respect to other planes. The observed evidence for a decrease of the blocking effect along this plane was a clear signature of fission times that are even larger than $10^{-17}$s [21].

These fission experiments were followed by the study of heavy compound nuclei ($Z_{proj} + Z_{crystal} \geq 114$) to infer the restauration of Coulomb barrier close to the expected stability-island of super-heavy nuclei, using low energy uranium and lead nuclei (CSS1 solo), and the INDRA multi-detector to detect all fission products, including light fragments. The results showed evidence for the existence of
long lifetimes \((t > 10^{-18}\text{s})\) for \(Z= 120\) and 124, but not for \(Z=114\) [22]. However, as shown in the quoted article, not all blocking dips could be reproduced by simulations because of the experimental resolution, and further investigations would be needed to show unambiguously that no long fission times are observed for other species.

3.4. Cooling and heating
In the late 90’s – early 2000’s, W. Assmann et al. have shown evidence for the breaking of the reversibility rule with low energy heavy ions, i.e. non uniform flux was found at emergence for uniform incidence with angular divergences large compared to critical channeling angles. Either cooling or heating of the transverse energy could be observed, depending on the ion energy, the ion and crystal atomic numbers, and the considered crystallographic direction. A large amount of experimental data was accumulated, among them some at GANIL in collaboration with CIMAP, and the interpretation was given in terms of impact parameter charge exchange and its influence on the conservation of transverse energy [23].

3.5. Ion guiding with micro-capillary
It has been shown that low energy charged ions can be guided through insulating pores or capillaries, by means of a dynamic charge patch formation at places where the incident particles hit the walls inside the capillaries [24]. Since then many groups explored this phenomenon that is closely related to channeling, although the guiding potential is not due to the static screened atomic potential. Indeed similar scaling laws were found as in channeling to describe the motion of guided ions. A collaboration between CIMAP, Riken and Kalamazoo studied these properties with glass capillaries of microscopic dimensions, using the low-energy beam lines of GANIL and multiply charged heavy ions [25].

4. Concluding remarks
The channeling and blocking phenomena offer unique techniques to perform multi-disciplinary studies with fast heavy ion beams, with applications both in nuclear physics (nuclear lifetime measurements, observation of rare Coulomb excitation modes like nuclear RCE or NEEC), atomic physics (detailed ion-electron interaction related charge-exchange processes like REC, RTE, EII), ion-solid interaction (impact parameter dependent energy loss, surface emission studies, surface and bulk damage). The high quality of stable beams and the high-resolution spectrometers and detectors available at GANIL, offer excellent opportunities to perform such experiments. The studies performed at GANIL are quite complementary to those performed at GSI, where higher energy beams are available, but also higher charge-states up to the heaviest ions at low energy after stripping and deceleration (see e.g. [26]). The study of charge exchange processes may have reached some limits, as well as nuclear physics studies, beyond which experiments become quite complex and require very long beam time. In the next future, application of micro-beams delivered through micro-capillaries at low energy may find applications in lithography and precise biological material irradiation. The medium- and low- energy highly charged ions of GANIL that correspond to the maximum of dose deposition in solids are still promising for new physical results, by using the selective energy loss associated to non-random trajectories inside crystal channels as performed in a SiO\(_2\) crystal [27]. In particular, surface and bulk damage studies could be performed under ultra-high vacuum. An equipped chamber is now available [28], but still a goniometer needs to be implemented in this UHV setup.

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