Swift heavy ion induced electron emission from solids

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Abstract. We briefly summarize the results of numerous experiments performed at GANIL aimed at measuring electron yields and doubly differential yields (energy or velocity spectra at different ejection angles, angular distributions). These studies, supported by theoretical investigations and numerical simulations, contributed decisively to our understanding of the very first step in energy deposition in matter, i.e. ionization and subsequent electron transport through condensed matter. The emitted electron spectrum contains a rich variety of features including binary encounter electrons (BEE), convoy electrons (CE), Auger electrons (AE) and the low-energy peak of “secondary” electrons (SE).

1. Electron Ejection by swift heavy ions at CIRIL-GANIL

Radiation effects in condensed matter emerge from “primary ionization” (and excitation) of target atoms, which leads to liberation of electrons. Electrons play thus a key role in track formation and in the related damage creation. A detailed knowledge of the structure of ion tracks is a key issue for our understanding of radiation effects in condensed matter (track formation, radiolysis, RBE calculations for hadrontherapy, etc.), where doubly differential electron ejection cross sections (DDCS) are a key input parameter for numerical simulations. Experimentally, electron yields, i.e. the mean number of emitted electrons, have been studied. Differential cross sections are measured with either magnetic or electrostatic spectrometers. Furthermore, the time-of-flight technique was used. GANIL is particularly well suited for this method, since it delivers pulsed beams with good time resolution (< 1 ns). The spectrometer is placed at a certain observation angle \( \theta \). The ejection angle \( \theta \) is usually measured with respect to the beam direction (\( \theta = 0 \) deg.). Often, thin foils are used as targets, which allows in addition to “backward” emission (\( \theta > 90 \) deg.) also to study electron ejection in “forward direction” (\( \theta < 90 \) deg.). Figure 1, taken from a review on achievements and open questions in electron ejection by swift ions [1], shows schematically some of the mechanisms involved.

Fast electrons induced by swift ions are often referred to as “\( \delta \)-rays” or “\( \delta \)-electrons”. The electrons kicked by the projectile may then undergo elastic or inelastic scattering during their travel through the medium (“electron transport”). The inelastic scattering liberates target electrons, thus creating in turn,
Figure 1. Schematic representation of mechanisms intervening in electron ejection from a thin solid foil of thickness $d$ bombarded with an ion beam of incoming charge $q_i$ and outgoing charge $q_f$. Primary ionization in close collisions leads to ejection of fast electrons from the target atoms ("binary encounter", denoted P, bottom part). Low energy electrons stem from ionization in distant collisions or collective effects (plasmon decay, wake, upper left part). On their way through the solid towards the surface, electrons suffer inelastic collisions without large angular deflections and angular scattering by the screened nuclei (upper right part). Also, an electron carrying projectile can be ionized by collisions with the target atoms (electron loss, denoted T, bottom part). Higher order collision sequences (P-T-P- ...) may occur ("Fermi shuttle", bottom part). The figure is taken from reference [1] with permission by the Royal Danish Academy of Science and Letters.
a cascade of “secondary electrons”. Finally, some of the electrons liberated inside the solid reach the surface and may cross the surface potential barrier to be ejected into vacuum as summarized in figure 1a). Consequently, the emitted electron spectrum contains a rich variety of features including binary 
encounter electrons (BEE) and convoy electrons (CE), Auger electrons (AE), see figure 2, and the low-energy peak of “secondary” electrons (SE) in the eV range. BEE electrons are ejected with a velocity of \( v_{\text{BEE}} = 2 v_p \cos(\theta) \) if the interaction with the target nucleus is neglected. For relativistic collisions, the momentum \( p = \gamma v \) with \( \gamma = (1-(v/c)^2)^{-1/2} \) is the relevant quantity instead of the projectile velocity \( v_p \). The observed BEE peak reflects the initial momentum distribution of the bound target electrons (“Compton profile”). CE stem from either electron loss or capture to low-lying projectile continuum states. Their energy is close to zero in the projectile frame, and they form a characteristic cusp shaped peak in fast forward electron spectra. Furthermore, target AE (typically at low energies \( E < 3 \text{ keV} \)), as well as projectile AE from in-flight de-excitation of heavy ions carrying electrons in bound excited states can be observed. Ejected electrons probe and provide information on interactions in different depths originating from different collision kinematics ranging from “soft” collisions and plasmon excitation to very “hard” close collisions. Most of the electrons have relatively low energy. We will -somewhat arbitrarily- consider “low energy electrons” as those that can typically be measured by charge collecting methods (for electron yields) or with a small electrostatic spectrometer (electron spectra, \( 0 \text{ eV} < E < 2 \text{ keV} \)).

The various experiments performed at GANIL aimed at measuring electron yields, and doubly differential yields (energy or velocity spectra at different ejection angles). They contributed decisively to our understanding of the very first step in energy deposition in matter, i.e. ionization and subsequent electron transport through condensed matter. One of the first experiments on electron emission from foils at GANIL showed that CE are very sensitive to the pre-equilibrium charge state evolution [2]. The emission of convoy (and BE-) electrons was also studied under channeling conditions [3]. More recently, new interest in electron emission under channeling conditions has arisen. The statistical distribution of the probability that \( n \) electrons are ejected \( P(n) \) (from which electron yields can be deduced) were measured in coincidence with the energy loss \( dE/dx \) and the emerging charge state of the projectiles [4]. Also, angular distributions of fast electrons were studied in connection with the detection of single heavy ions with CCD detectors [5] being used in radiobiological experiments.

### 2. Slow electrons

The first systematic studies of electron emission at CIRIL and GANIL were measurements of backward and forward electron yields as a function of carbon foil thickness [6-8]. These experiments allowed to establish the behaviour of electron emission as a function of the projectile velocity \( v_p \), the projectile atomic number \( Z_p \) and target thickness \( d \). Deduced simple scaling laws are important for applications, where often thin foils are used as electron source for e.g. beam monitors and dosimetry. These properties and scaling laws are discussed in [1]. Furthermore, the experimental data served as “benchmark” for numerical simulations of primary ionization and electron transport in solids [6, 7].

Low energy electron emission strongly depends on surface properties [9]. Therefore, experiments with sputter-cleaned surfaces in ultra-high vacuum are desirable and a specific set-up was built at CIRIL [10]. Electron energy spectra \( E < 1 \text{ keV} \) and simultaneously measured electron yields were studied as a function of the projectile charge \( q_i \) at fixed velocity \( 9 \text{ MeV/u} \) [10-13]. Important results on “high charge effects” related to the strong perturbation in heavy ion collisions were obtained from those measurements and comparison to theoretical calculations of low energy electron spectra. The perturbation strength was varied from \( q/v_p = 0.002 \) (very weak perturbation, protons) to \( q/v_p = 2 \) (strong perturbation, GANIL heavy ions). The experimental results show a “reduction effect” with respect to a \( q^2 \) scaling for low energy electron emission under strong perturbation by heavy projectiles [10-12]. Such effects could arise from either specific high charge effects in primary ionization, or collective effects on electron transport in the vicinity of the projectile due to positively charged zone in the ion wake, which might attract the electrons moving away from the ion track [12, 14]. Theory rather points toward saturation of primary ionization cross sections with increasing ion charge [12, 15-16].
Figure 2. Fast forward electron spectra (the doubly differential electron yield) at small ejection angle \( \theta = 2^\circ \) close to the beam direction as a function of the electron time of flight from impact of \( ^{78}\text{Kr}^{32+} \) and \( ^{78}\text{Kr}^{34+} \) (64 MeV/u) on a thin carbon foil. Prominent peaks are those of convoy electrons (CE, which with electron carrying projectiles mainly stem from projectile electron loss, denoted “T” in figure 1b) at longer time-of-flight (TOF) and of binary encounter electrons (BEE, denoted “P” in figure 1b) at shorter TOF. With \(^{78}\text{Kr}^{32+}\) also Auger electrons (AE) from the in-flight de-excitation of the excited projectile are observed.

Another specific feature observed with heavy ions is the appearance of Auger hypersatellite lines from double ionization of the carbon K-Shell [11, 17]. From a careful analysis of the shape of carbon KVV Auger spectra, including background subtraction and deconvolution techniques, an increase of the width of the primary Auger electron spectrum with ion charge is observed. This effect is strongly connected to a modification of the electronic band structure of the solid target at the moment of the Auger decay. By adjusting the temperature-dependent Fermi distribution folded with the density of states to the observed primary line width, a mean “electronic temperature” in the track can be deduced [13, 17-19]. Measured “track temperatures” can thus test the predictions of thermal spike numerical simulations [17, 19] and serve as an in-site thermometer of the transient hot “plasma” inside the track. This experimental method gives quantitative information about the onset of the thermal spike. Information about the “track temperature” at later times and its evolution may be accessible via the measurement of the velocity of sputtered particles [20].

3. Fast electrons

Besides the above mentioned studies of CE, BE and \( \delta \)-electrons [2, 3, 5] with the high energy GANIL beams, CE and BE were also studied at the medium energy facility SME [21, 22]. Also, by employing the multidetector array ARGOS, initially developed for studying nuclear reactions, and now adapted for detection of electrons (E > 10 keV) [23-27], ejection mechanisms of electrons at relativistic energies were studied in considerable detail. In comparison to conventional spectroscopic methods used in atomic collisions (i.e. electrostatic and magnetic analyzers), three important advantages were achieved: i) DDCS (velocity spectra) can be measured simultaneously at many ejection angles, ii) absolute cross sections can be measured with great accuracy thus allowing a stringent test of ionization theory iii) (multi-) coincidences can be measured. This latter feature opens the door for studies of processes which have not
yet been accessible up to now (e.g. correlated emission of two or more electrons). By focusing on high energy BEE, a relativistic generalization of the electron impact approximation (EIA) and of free-electron transport could be successfully performed [22-25]. Measurements of electron spectra as a function of the target thickness allowed observing how electron emission evolves from single collisions (as in atomic collisions with low density gas targets) up to multiple collisions (as in the bulk of solids, where electron transport phenomena become important) and thus to link single collisions to effects in condensed matter as shown in figure 1a) [22-23, 25].

The study of high-energy electrons well beyond the binary encounter peak by the CIRIL group has received considerable international attention. A particularly interesting result is the observation of unexpectedly high cross sections for electron emission at energies far beyond the BE peak. Possibly, this can be explained by the so-called "Fermi-shuttle" process, which was introduced to explain the origin of high-energy cosmic radiation due to acceleration of charged particles by repeated collisions with moving magnetic fields in interstellar space. Multiple scattering sequences of electrons on the combined projectile-target systems can lead to electron acceleration (figure 1b) and emission of fast electrons even at large (backward) emission angles [28, 29].

Ongoing work concerns the application of electron spectroscopy in collisions of swift ions with insulating targets. The slowing down of BEE and CE allowed determining the target material dependent charging-up of polymer foils [30]. The angular dependence of in-flight emitted projectile Auger electrons with Kr$^{32+}$ (64 MeV/u) [27] was also studied. For this collision system, the unique feature of the ARGOS multidetector, i.e. measurement of coincident emission of particles, allowed for the first time to study also the correlated emission of electrons. Under particular kinematic conditions (in-plane coincidence), a peak is observed which corresponds to an inelastic binary encounter with momentum transfer to an inner shell projectile electron. This is a rare ionization process with a specific kinematic signature involving a target and a projectile electron simultaneously, related to so-called (e,2e) experiments (one incoming electron, two outgoing electrons) [31]. Measurements of electron-X-ray coincidences are planned to study Radiative Electron Capture to the Continuum [32-33].

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References
[1] Rothard H and Gervais B 2006 Electron emission from solids irradiated with swift ion beams Ion Beam Science - Solved and Unsolved Problems (Mat. Fys. Medd. Dan. Vid. Selsk. 52 497-524) ed P Sigmund
[2] J. P. Gibbons J P, Elston S B, Kimura K, DeSerio R, Sellin I A, Burgdörfer J, Grandin J P, Cassimi A, Husson X, Liljeby L and Druetta M 1991 Observation of rapid evolution of convoy electron angular distributions Phys. Rev. Lett. 67 481–484
[3] Andriamonje S, Chevallier M, Cohen C , Dural J, Genre R, Girard Y , Groeneveld K O, Kemmler J, Kirsch R, L’Hoir A, Maier R, Poizat J C, Quéré Y, Remillieux J, Schmaus D and Toulemonde M 1991 Charge exchange processes of high energy heavy ions channeled in crystals Proceedings of the 4th Workshop on High Energy Ion-Atom-Collision Processes (Springer Lecture Notes in Physics vol 376) ed D Berényi and G Kock pp 221-234
[4] Barrué F, Chevallier M, Dauvergne D, Kirsch R, Poizat J C, Ray C, Adoui L, Cassimi A, Rothard H, Toulemonde T, Cohen C, L’Hoir A, Vernhet D, Demonchy C, Giot L, Mittig W, Pita S, Roussel-Chomaz P and Billebaud A 2004 Electron emission induced by fast heavy ions in a thin silicon crystal Phys. Rev. A 70 032902
[5] Meier MM, Schott J U, Strauch K and Groeneveld K O 1998 Detection of 8-electron events in CCD: a fingerprint of single swift heavy ions *Nucl. Instrum. Meth. Phys. Res. B* **146** 601-606

[6] Rothard H, Caraby C, Cassimi A, Gervais B, Grandin J P, Jardin P, Jung M, Billebaud A, Chevallier M, Groeneveld K O and Maier R 1995 Target thickness dependent electron emission from carbon foils bombarded with swift highly charged heavy ions *Phys. Rev. A* **51** 3066-79

[7] Jung M, Rothard H, Gervais B, Grandin J P, Clouvas A and Wünsch R 1996 Transport of electrons induced by highly charged Ni (74 MeV/u) and Cu (9.6 MeV/u) ions in carbon: a study of target thickness dependent electron yields *Phys. Rev. A* **54** 4153-4161

[8] Rothard H, Jung M, Caron M, Grandin J P, Gervais B, Billebaud A, Clouvas A and Wünsch R 1998 Strong projectile dependent forward/backward asymmetry of electron ejection by swift heavy ions in solids *Phys. Rev. A* **57** 3660-64

[9] Caron M, Rothard H and Clouvas A 2003 Influence of Adsorbates on Electron Emission from amorphous Carbon under Electron and swift heavy Ion Bombardment *Eur. Phys. J. Appl. Phys.* **21** 9-16

[10] Caron M, Rothard H, Jung M, Mouton V, Lelièvre D, Beuve M and Gervais B 1998 Mobile UHV Set-up for the Investigation of Electron Spectra at large Accelerators: first Results on High Charge Effects *Nucl. Instrum. Meth. B* **146** 126-130

[11] Caron M, Rothard H, Beuve M and Gervais B 1999 Multiple ionization and high charge effects in electron spectra from impact of 9 MeV/u ions on carbon *Phys. Scripta T* **80** 331

[12] Beuve M, Caron M, Gervais B and Rothard H 2000 Charge dependence of electron emission in swift heavy ion collisions with carbon *Phys. Rev. B* **62** 8818-8823

[13] Caron M, Rothard H, Beuve M and Gervais B 2001 Shape analysis of Auger electron spectra induced by highly charged ion impact on carbon *Phys. Scripta T* **92** 281-283

[14] Schiwietz G, Grande P, Skogvall B, Biersack J P, Köhrbrück R, Sommer K, Schmoldt A, Goppelt P, Kadar I, Ricz S and Stettner U 1992 Influence of nuclear track potentials in insulators on the emission of target Auger electrons *Phys. Rev. Lett.* **69**, 628-631

[15] Beuve M, Caron M, Fainstein P D, Galassi M, Gervais B, Rivarola R D and Rothard H 2002 Monte Carlo simulation of electron emission induced by swift highly charged ions: beyond the linear response approximation *Eur. Phys. J. D* **21** 125-135

[16] Gervais B, Beuve M, Caron M and Rothard H 2003 Saturation effects in highly charged ion interaction with thin carbon foils *Nucl. Instrum. Meth. Phys. Res. B* **205** 835-840

[17] Schiwietz G, Xiao G, Luderer E and Grande P L 2000 Auger electrons from ion tracks *Eur. Phys. J. D* **21** 1563-1578

[18] Schiwietz G, Xiao G, Grande P L, Luderer E, Pazirandeh R and Stettner U 1999 Determination of the electron temperature in the thermal spike of amorphous carbon *Europhys. Lett.* **47** 384-390

[19] Caron M, Rothard H, Toulemonde M, Beuve M and Gervais B 2006 Theoretical and experimental etudy of electronic temperatures in heavy ion tracks from Auger electron spectra and thermal spike calculations *Nucl. Instrum. Meth. B* **245** 36-46

[20] Hijazi H, Rothard H, Boduch P, Alzaher I, Langlinay T, Cassimi A, Ropars F, Been T, Ramillon J-M, Lebius H, Ban-d'Etat D, Farenzena L S and da Silveira E F 2012 Electronic sputtering of LiF by Krypton (10 MeV/u): size dependent energy distributions of Li+(LiF)n clusters *Eur. Phys. J. D* **66**: 305

[21] Rothard H, Jakubassa-Amundsen D H and Billebaud A 1998 Solid State Effects in Binary Encounter Electron Emission from 13.6 MeV/u Ar17+ Collisions with C, Al, Cu and Au Fools *J. Phys. B* **31** 1563-1578

[22] Jakubassa-Amundsen D H and Rothard H 1999 Fast Electrons from Collisions of highly stripped Ions with solid-state Targets *Phys. Rev. A* **60** 385-391
[23] Rothard H, Lanzanò G, Jakubassa-Amundsen D H, De Filippo E, Mahboub D 2001 Theory and Measurement of absolute doubly differential Cross Sections of Binary Encounter Electron Ejection in Collisions of swift heavy Ions with Solids J. Phys. B 43 3261-3274

[24] Lanzanò G, De Filippo E, Aiello S, Geraci M, Pagano A, Cavallaro S, LoPiano F, Pollaco E C, Volant C, Vuillier S, Beck C, Mahboub D, Nouicer R, Politi G, Rothard H and Jakubassa-Amundsen D H 1998 Fast Electron Production in Atomic Collisions induced by 77 A-MeV Ar Ions studied with a Multidetector Phys. Rev. A 58 3634-3641

[25] De Filippo E, Lanzanò G, Rothard H, Volant C, Jakubassa-Amundsen D H, Aiello S, Anzalone A, Arena N, Geraci M, Giustolisi F and Pagano A (2003): Absolute Cross Sections for Binary-Encounter Electron Ejection by 95 MeV/u 36Ar 18+ penetrating Carbon Foils Phys. Rev. A 68 024701

[26] De Filippo E, Lanzanò G, Rothard H, Volant C, Aiello S, Anzalone A, Arena N, Geraci M, Giustolisi F, and Pagano A 2004 Fast Electron Ejection from C, Ni, Ag and Au Foils by 36Ar 18+ (95 MeV/u): Measurement of absolute Cross Sections Eur. Phys. J. A 21169-174

[27] Lanzanò G, De Filippo E, Hagmann S, Rothard H and Volant C 2007 In flight emission of projectile Auger electrons from highly energetic heavy ions (20 MeV/u<E<100 MeV/u) colliding with carbon foils Nucl. Instrum. Meth. B 256 510-514

[28] Sulik B, Stolterfoht N, Hellhammer R, Pésic Z, Koncz C, Tőkési K and Berényi D 2003 Fermi-shuttle acceleration of electrons in ion–matter interaction Nucl. Instrum. Meth. B 212 33-44

[29] Rothard H, Lanzanò G, De Filippo E and Volant C 2005 Fermi Shuttle Acceleration in atomic Collisions: the Case of ion induced Electron Emission Nucl. Instrum. Meth. B 230 419-425

[30] De Filippo E, Lanzano G, Amorini F, Geraci E, Grassi L, La Guidara E, Lombardo I, Politi G, Rizzo F, Russotto P, Volant C, Hagmann S and Rothard H 2011 Slowing down of fast electrons as probe for charging and discharging dynamics of ion-irradiated insulators Phys. Rev. A 83 064901

[31] De Filippo E, Lanzanò G and Rothard H 2007 Impact and Applications: Fast Electron Ejection in Atomic Collisions Studied with the ARGOS Multi-Detector Nucl. Phys. News. 17 No. 2 24-27

[32] Hillenbrand P M, Hagmann S, Stoehlker T, Litvinov Y, Kozhuharov C, Spellmann U, Shabev V M, Stiebing K, Leitschinsky M, Surzhkyov A, Voitkiv A, Franzke B, Fischer D, C. Brandau C, Schippers C S, Mueller A, Schneider D, Jakubassa D, Artiomov A, De Filippo E, Ma X, Dörner R and Rothard H 2013 Future experiments using forward electron spectroscopy to study quantum dynamics of high-Z ions at the ESR/CRYRING storage rings Phys. Scripta. T 156 014087

[33] Hillenbrand P M, Hagmann S, Atanasov D, Banaš D, Blumenhagen K-H , Brandau C, Chen W, De Filippo E, Gumberidze A, Guo D L, Jakubassa-Amundsen D H, Kovtun O, Kozhuharov C, Leitschinsky M, Litvinov Yu A, Müller A, Müller R A, Rothard H, Schippers S, Schöffler M S, Spellmann U, Surzhkyov A, Trotsenko S, Winckler N, Yan X L, Yerokhin V A, Zhu X L and Stöhlker T 2014 Radiative-electron-capture-to-continuum cusp in U88+i+N2 collisions and the high-energy endpoint of electron-nucleus bremsstrahlung Phys. Rev. A 90 022707