GSI-ESAC/Materials Research

Date: 13 April 2004

Letter of Intent

for

Materials Research with Relativistic Heavy Ion Beams

Abstract

This Letter of Intent proposes the study primarily of two subjects: *(1) Heavy ion-induced modifications of solids that are exposed to extremely high pressures.* For this purpose, a solid must be enclosed between anvils, which cause this pressure. Therefore, the ions require high kinetic energy to penetrate through one of the anvils, reach the sample, and experience there maximum energy loss (Bragg peak) or, alternatively, pass fully through still with relativistic velocity. In addition to pressure, also other parameters such as temperature and shear forces influence the processes. The heavy ions deposit large amounts of energy in a very short time and within a very small volume, and in this way act as a trigger for alterations already near to a threshold. Phase transitions and acoustic wave propagation, the latter being preferably stimulated by short, intense ion pulses in macroscopic samples, are important aspects on which efforts will focus. *(2) Analysis of material modifications induced by relativistic heavy ions.* The short-time signature of the processes stimulated by the passage of the projectiles comprises electrons, ions, clusters, and electromagnetic radiation (such as X-rays and Cerenkov light) emitted from the ion trajectories. Their properties concerning intensity, energy, time scale, and spatial distribution provide insight in the formation of tracks. This information will be complemented by the analysis of the final material modifications.

GSI-ESAC/Materials Research

Date: 08 April 2004

Letter of Intent

for:

Materials Research with Relativistic Heavy Ion Beams

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1. Introduction

Energetic heavy ions traveling through a solid exert an enormous electric field (of order 10^{12} V/m) over an extremely short time period (some 10^{-18} s or even less) at the site of those constituents (atoms, electrons, etc.) of the solid that are in close proximity to the trajectory, thereby successively transferring their kinetic energy to the solid. This energy loss triggers complex, rapid processes, beginning with the generation of fast electrons, followed by Coulomb repulsion of the ionized atoms, by X-ray and Auger electron emission, electron-phonon coupling, etc. As a final result, the projectiles may cause long, narrow modified zones with radial extensions of a few nanometers. Due to their macroscopic length, they are quasi-one-dimensional perturbations of the solid. In many materials, dramatic modifications are induced inside the cylinder-shaped tracks such as changes from crystalline to amorphous and the creation of metastable high-temperature or high-pressure phases. The track features constitute signatures of the specific properties of a given solid and reveal, in which way this solid copes with the deposition of energy under extreme conditions [SHIM02].

Modifications induced by ions of a total kinetic energy up to several GeV are one of the subjects permanently studied in materials research. There exists a large amount of experimental data for different classes of solids, such as polymers, metals, ionic crystals, and some selected semiconductors, to name a few. The alterations have been analyzed with a variety of methods including high-resolution microscopy, optical spectroscopy, X-ray and neutron scattering, etching, nanoprofilometry, Rutherford backscattering, and others. Furthermore, the experimental findings were accompanied by the development of theoretical concepts concerning the mechanisms of track creation. The thermal spike and the pressure pulse model may be mentioned here as prominent examples.

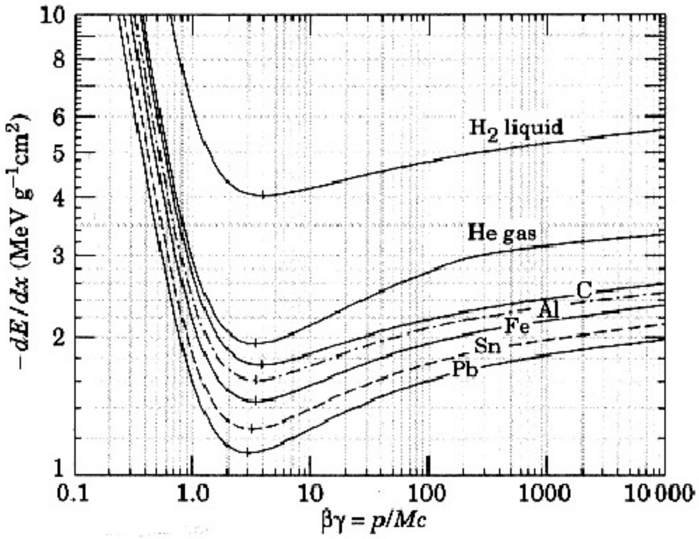

Fig. 1: Energy loss dE/dx of protons in matter as a function of their velocity.

The most important quantity for track formation is the stopping power d*E*/d*x* (energy loss per unit path length) of the projectile. The mechanisms of d*E*/d*x* are rather well understood for non-relativistic velocities. As the velocity increases to relativistic values, d*E*/d*x* decreases reaching a minimum at about 2 GeV/u. For even higher ion energies, new phenomena set in due to relativistic effects, and the stopping power slightly increases again (Fig. 1). Comprehensive experimental d*E*/d*x* studies have been performed at projectile energies in the range ≤ 1 GeV/u [Sc94, Sc98, Ge98, We00, Mo96, Do98, **Geissel2003**]. In contrast to that, only scarce quantitative experimental d*E*/d*x* results are presently available above 1 GeV/u (a single data point existing for 160 GeV/u Pb ions [Da96, Da00]). The understanding of basic energy loss processes at the extremely high beam energies [Li96, Sh00] must be improved by new experiments using different kinds of materials.

Besides the size of d*E*/d*x*, the energy density plays a crucial role for track creation. For higher ion velocities, energetic knock-on electrons distribute their energy radially in a larger cylindrical volume. Therefore, the energy density per atom (or energy per lattice plane) decreases (Fig. 2). Concerning material modifications, the energy density effect of such energetic ion projectiles is not known. At ultrahigh ion energies, tracks in very sensitive materials (e.g., polymers) may be enlarged, whereas in resistant materials, the critical energy density is possibly not surpassed and track formation is suppressed.

While being exposed to energetic heavy ions, solids may simultaneously be put under high pressure and kept at a low or elevated temperature. This supports the goal to reach new phases or to test the stability of a material far from ambient conditions. Very promising experimental possibilities arise for the geosciences. It should also be emphasized that there exist links and fluid transition between materials research and plasma physics.

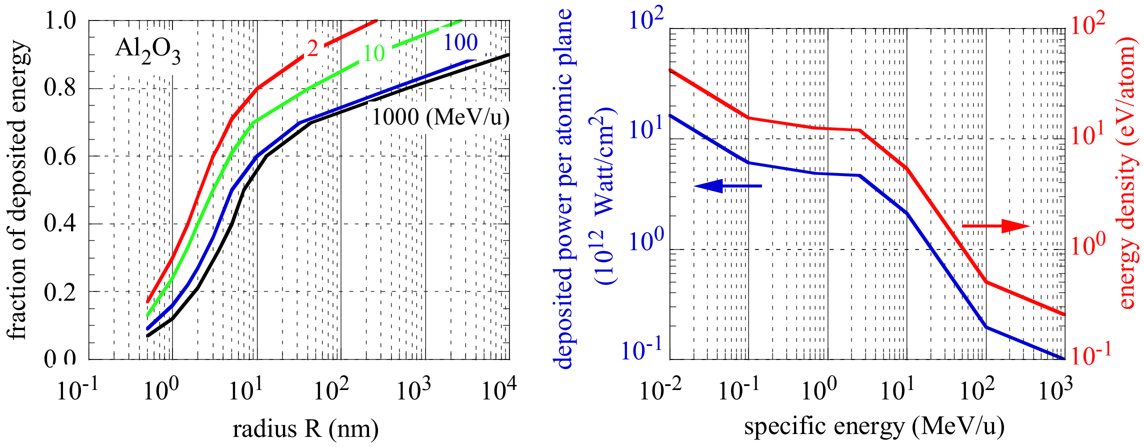

Fig. 2: Effect of ion velocity on radial energy density
Left: Fraction of energy deposited in a cylindrical region of radius R for low and high ion energies.
Right: Energy density (eV per atom) and power transfer per lattice plane (Watt/cm^2) versus energy.

Based on the high energies and small stopping powers, the new ion beams have much larger penetration depths. Novel types of experiments will become possible. This Letter of Intent comprizes two subjects: *(1) Heavy ion-induced modifications of solids that are exposed to extremely high pressures* and *(2) Analysis of material modifications induced by relativistic heavy ions.* These objectives are addressed in the following sections.

2. Science Case

2.1. Ion-induced modifications of pressurized solids

The goal is to study modifications that solids, being already exposed to a very high pressure, undergo if energetic heavy ions pass through or are stopped in the sample under study. To realize such investigations, two indispensable conditions must be fulfilled: First, such a solid must be enclosed between anvils, which cause the pressure. Second, as a consequence, the ions require high kinetic energy to penetrate through one of the anvils and reach the sample. When aiming at large alterations of a solid, one would

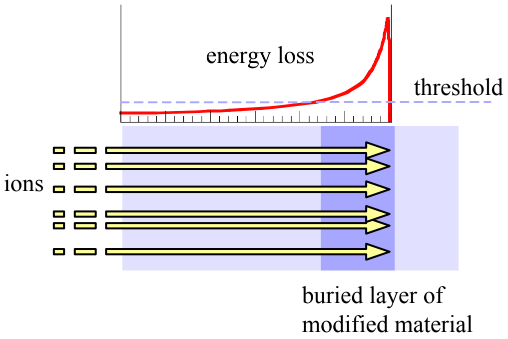

Fig. 3: Material modification by locating the Bragg peak inside the pressurized sample.

locate the so-called Bragg peak, representing maximum stopping power, inside the sample (see Fig. 3). If the projectiles have to pass anvils with lengths of many cm, then the ion energy must be in the regime of several GeV/nucleon and thus significantly beyond the energies provided by SIS.

The heavy ions deposit during a very short time a large amount of energy within a very small volume. In this way, they can act as a trigger for alterations of the structure of the solid, which already may be near to an energy threshold, from which upward these structure changes will occur. The combination of high pressure and very fast, intense, and locally confined energy supply, provided by energetic heavy ions, opens up the possibility of producing new phases not realizable with pressure alone.

A further aspect of great interest is the creation of acoustic waves, stimulated by short, intense ion pulses in pressurized samples. In order to study wave propagation over reasonably large distances, the samples require mm or even cm sizes, and again voluminous anvils must be penetrated by the ion beam. Ultrasonic waves induced by short ion pulses in solids under ambient pressure have already been studied in detail [KK00, KK02]. In addition to pressure, there are other parameters that influence the processes, in particular temperature, shear forces, and ion- and solid-specific energy losses. Given, besides these parameters, the large spectrum of different solids, a realm of new phenomena must be expected.

The exposure of pressurized solids to radiation is commonly used for analysis by means of X-ray and neutron diffraction, and optical (such as e.g. Raman) spectroscopy. Also for analysis purposes, muons were stopped in a solid under high pressure combined with muon spin resonance measurements [SG97]. In both cases, the radiation serves as a probe, and a modification of the sample is not desired. An early example of intended sample changes is gamma irradiation of polymers at high pressure [MK86]. The influence of stress on the formation of ion tracks in amorphous iron boron alloy was demonstrated by causing a stress zone with a first heavy-ion irradiation and subsequently exposing the sample including this zone to a second irradiation. The tracks in the stress zone were less pronounced than those in previously non-irradiated areas [TK00].

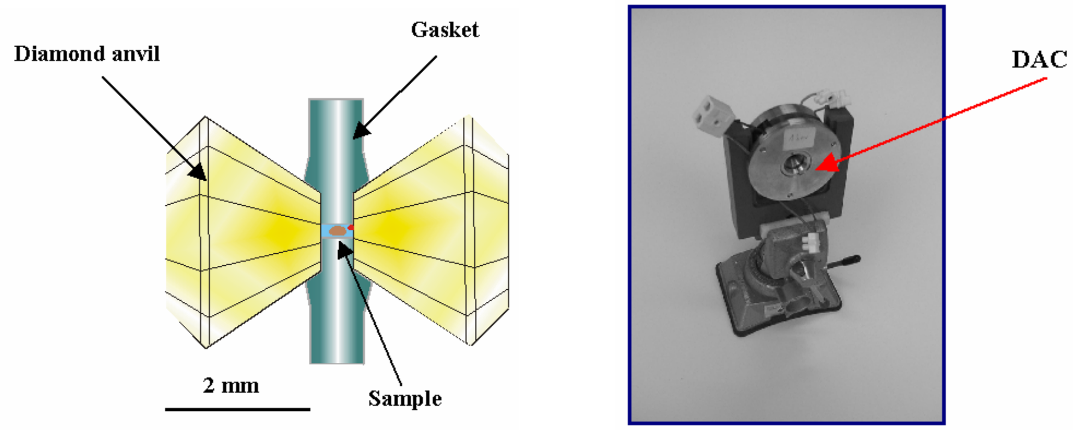

Fig. 4: Scheme and photograph of a diamond anvil cell (DAC) used for heavy-ion irradiation [LG03, GL04].

A series of irradiations of solids pressurized in diamond anvil cells have been performed in the recent past with beams from the SIS heavy-ion synchrotron at GSI [LG03, GL04]. These experiments aimed to gain first experience, in which way solids at high pressure react to the passage of energetic heavy ions. This work was originally stimulated by the great relevance of pressure in the field of geochronology and geophysics where tracks of fission fragments formed in pressurized rocks are commonly used for dating [Re00]. Small samples of phlogopite (a natural dark mica mineral) and highly oriented pyrolytic graphite (HOPG) were exposed to a maximum pressure of 2.2 and 12.1 GPa, respectively, in a diamond anvil cell, and were irradiated with highly charged uranium and gold ions (fluences about 1×10^{11} ions per cm^2). The projectiles had primary kinetic energies between 200 and 400 MeV/nucleon, necessary for passing one of the two diamond anvils with typical lengths of order 2 to 4 mm. The primary ion energy was chosen such that highest energy loss (Bragg peak) occurred inside the sample under investigation. Subsequent etching showed that, despite the enormous pressure, each ion caused a latent track in phlogopite. In HOPG, TEM images visualized amorphized zones containing small regions of recrystallized graphite.

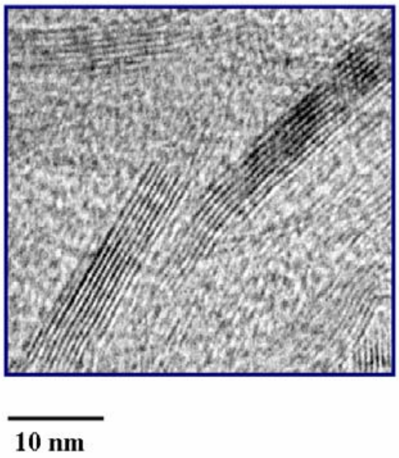

Fig. 5: TEM micrograph of HOPG irradiated with gold ions of primary SIS energy 400 MeV/u under a hydrostatic pressure of 8.5 GPa. Recrystallized graphite stripes embedded in an amorphous matrix are visible.

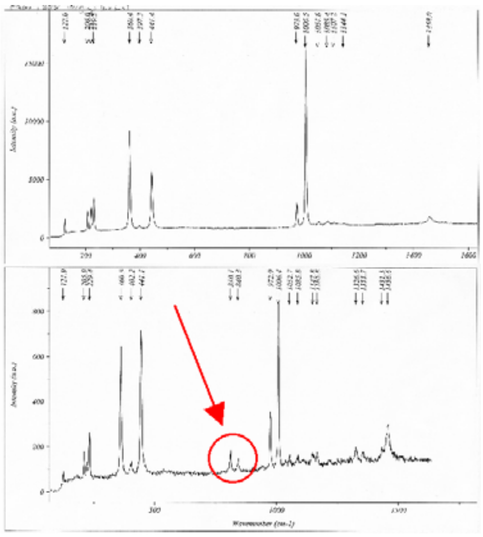

Fig. 6: Raman spectra of zircon before (top) and after (bottom) irradiation with uranium ions of primary SIS energy 200 MeV/u. The interaction of the heavy ions with the zircon sample under 14.1 GPa leads to the creation of reidite, a polymorphous high- pressure phase.

2.2. Analysis of material modifications induced by relativistic heavy ions

For a better understanding of the track formation mechanisms, it is extremely important to investigate transient processes taking place during or shortly after the passage of the ion projectile (Fig. 7). Electronic processes dominate the track evolution on a time scale from roughly 10^{-18} to 10^{-13} s. The dynamics of damage production on the scale in the order of 10^{-14} s can be studied by high-resolution convoy- and Auger-electron spectroscopy [Sc92, Xi97] as well as by X-ray detection. However, these short-time techniques yield information on the heating-up of the electrons in the bands before the atoms are set in motion and therefore do not give a complete picture of the track formation mechanisms. The displacement of atoms resulting from the high electronic excitation typically occurs on a time scale of 10^{-13} to 10^{-12} s, but also includes slower processes related to the cooling of the lattice or, for example, on shock wave propagation. The creation of a shock wave as a consequence of the projectile passage can be studied in solid targets by monitoring ultrasonic signals (pulse width between 3 and 600 ns) by means of piezoelectric sensors [KK00, KK02].

The beam parameters of the new facility, in particular the combination of much higher intensities and shorter ion pulses will allow an access to fast processes. Such experiments require pulse length of 10^{-11} s or shorter and a high temporal resolution of signal diagnosis. Extremely rapid processes may become possibly accessible by collecting Cerenkov light produced by ultrarelativistic ions. The collected light is auto-synchronized with the transient track processes and can be used to directly probe modifications induced in the solid by monitoring transient changes of transmission, reflectivity, or scattering of diffuse light on the target surface similar to pump-probe experiments with picosecond and femtosecond laser pulses. But also on a much longer time scale (10^{-9} s), interesting processes take place in specific materials, such as for example radical reactions in polymers (see Fig. 7).

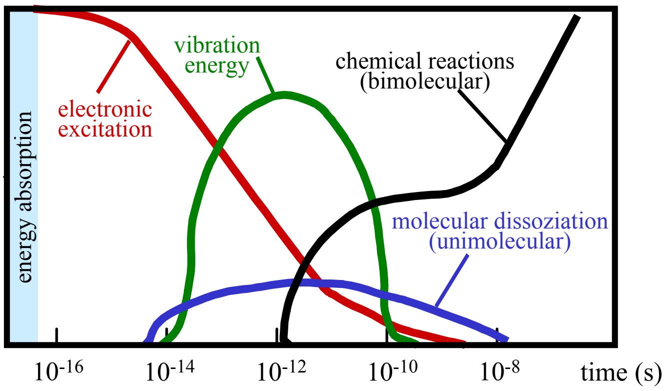

Fig. 7: Schematic of energy dissipation versus time in polymers after ion irradiation.

3. Facilities and Instrumentation

3.1. Demands on experimental area and ion-beam characteristics

Fig. 8 depicts the experimental area for atomic physics, materials research, and biophysics, as displayed in the FAIR Conceptional Design Report. This scheme is considered as adequate for the aims pursued by materials research.

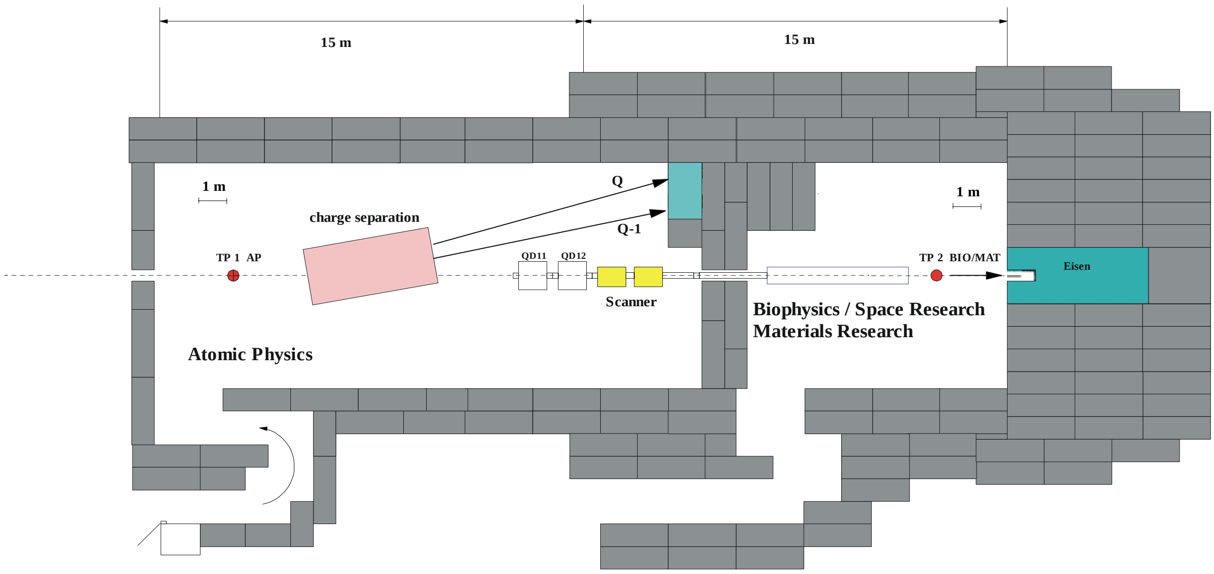

Fig. 8: Experimental area for atomic physics, materials research, and biophysics using ion beams from SIS-18 and SIS-100.

An indispensable requirement is a sufficient amount of space behind the beamline, which means the following: There must be room for the installation of an ultrahigh pressure apparatus with large pressure anvils (see Fig. 9). Such a machine may need of order 4 m^2 for installation on the floor and about 3 m free height. Additionional space must be available for common power supply equipment and for analytical instrumentation as, for example, optical spectrometers, ion detectors, etc. An area of 10 m^2 is therefore regarded as a minimum.

Materials research intends to exploit even the highest kinetic energies and the heaviest ion species provided by FAIR. For studies of the full penetration depth of such projectiles in various solids, particularly also in polymers, where the range will be large due to the low density, pieces of solids with lengths of 1 to 2 m will be mounted behind the beamline exit window.

3.2. High-pressure arrangement

A modern ultrahigh-pressure apparatus equipped with a large-volume multi-anvil cell is depicted in Fig. 9. This type of arrangement allows the mounting of samples of several cm^3. Volumes of that size make it possible to study on a macroscopic scale the propagation of ultrasonic waves induced by short heavy-ion pulses.

Fig. 9: Ultrahigh-pressure facility with large-volume multi-anvil cell (taken from the homepage of Bayerisches Geoinstitut, Bayerische Akademie der Wissenschaften, Bayreuth).

3.3. Detection of particles and radiation emitted from a target

The investigation of energy loss mechanisms of relativistic heavy ions in solids requires the mounting of samples of considerable length, that means 1 to 2 m for ions in the multi-GeV/nucleon range. The full penetration depth and the material modifications will be investigated off-line subsequently. Nevertheless, the apparatus must be equipped with detectors for electrons, ions, atoms, clusters, and electromagnetic radiation (such as X-rays and Cerenkov light), which are emitted from a narrow cylindrical zone around the trajectory of each single ion. Time-resolving techniques (or example convoy and Auger electron spectroscopy) should be available to access the time scale $< 10^{-13}$ s.

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