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Low-energy μSR investigations of photo-induced effects on a nm scale

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Abstract

Photogenerated charge carriers open the fascinating possibility to tune charge carrier concentrations in situ, which provides a convenient way to change physical properties of a system. Low-energy μSR is ideally suited for this kind of studies due to the perfect overlap of muon stopping distribution and the region of highest photon absorption close to the surface. We present a description of the current experimental setup and the results of a prototype experiment in Si. These results demonstrate the very promising prospect of extending LE-μSR to the investigation of photo-induced effects on a nanometer scale near surfaces, in thin films and heterostructures in general.

Keywords: low-energy muons, low-energy μSR (LE-μSR), photo-induced effects, nanometer scale investigations

In a large variety of physical systems small changes in charge carrier densities may induce dramatic changes of their physical properties. Prominent examples are complex transition metal oxides, including cuprates and manganites, representing strongly correlated electron systems close to a metal-insulator transition. Photogenerated charge carriers are of particular interest for the development of new functional devices for optoelectronic or spintronic applications, like photoswitches, phototransistors or photomemories. Here, the focus is on thin films and interfaces, with the goal to tailor novel properties and behaviour. A local probe such as the μ^+ can provide complementary and potentially new information to macroscopic measurements, due to its high sensitivity to the local magnetic and electronic environment. As a local spin-1/2 probe "sitting" at an interstitial lattice location it is a very versatile tool to measure internal magnetic fields and magnetic field fluctuations to probe phase transitions, spin dynamics, magnetic penetration depths in superconductors, hydrogen-like muonium (Mu) impurity states in semiconductors, and local environment and dynamics in molecular systems. The application of the μSR technique to study photo-induced effects offers very promising perspectives to gain new insights and results. For these studies a low-energy muon beam with tunable implantation energies < 30 keV (< 200 nm depth) is best suited, since many of the interesting systems consist of thin films or heterostructures with thicknesses of less than 100 nm. Another advantage of using low-energy muons is the matching between muon stopping site and region of maximum light absorption. Light penetrating matter is exponentially attenuated: most of it is absorbed within the first few hundred nm close to the surface, where the highest charge carrier concentration is also generated. Only in semiconductors with indirect band gap (e.g. Si, Ge) light with energy slightly larger than the band gap can generate photo-carriers in the sub-mm range due to the small absorption coefficient. Near the surface the charge carrier concentration can be varied not only by tuning the light intensity, but also by changing its energy, because the photon absorption coefficient strongly depends on photon energy.

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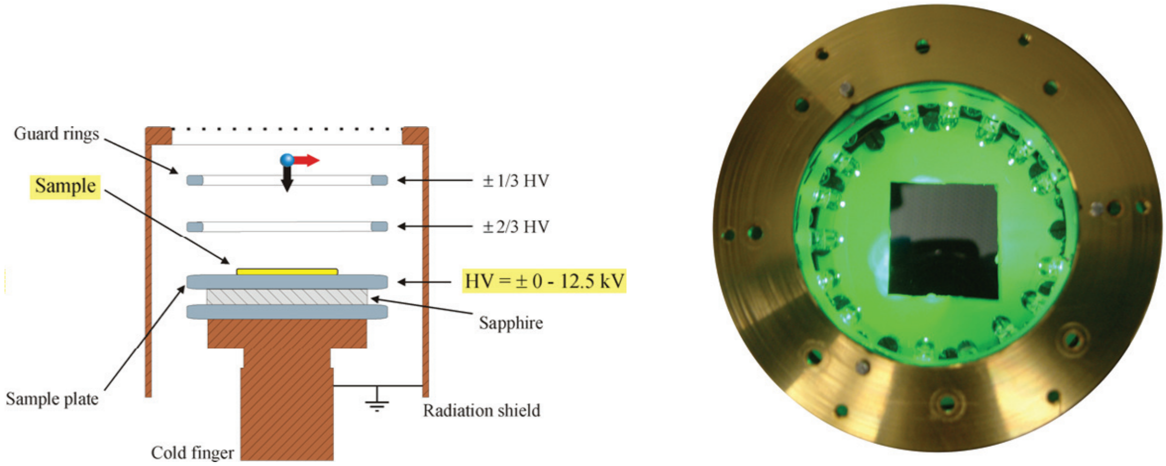

Figure 1: Left: sketch of the LE-μSR sample cryostat. The sample is electrically insulated from the cold finger of the cryostat by a sapphire disk. This allows to apply up to ± 12.5 kV on the sample to adjust the implantation energy. Two guard rings in front of the sample can be used to obtain a better homogeneity of the electric field. Right: view in beam direction onto the opening of the gold plated radiation shield of the LE-μSR setup, with a ring of 33 green LEDs, mounted inside the shield, illuminating a 3×3 cm^2 Si wafer. The muon beam size is about 2×2 cm^2. For installation of the LED ring the two guard rings in a) have been removed.

Up to now surface muon beams have been used for illumination studies on semiconductors (Si, Ge, GaAs [1, 2, 3, 4, 5, 6, 7]). The charge state and dynamics of Mu states in semiconductors are very sensitive to doping, i.e. the availability of free charge carriers [8, 9]. For example, in undoped Si two neutral paramagnetic Mu states exist at the tetrahedral interstitial site ($\mathrm{Mu_T^0}$) and at the bond center between two Si atoms ($\mathrm{Mu_{BC}^0}$). These and the charged diamagnetic state $\mathrm{Mu_{BC}^+}$ are distinguishable by their different precession signatures in transverse field (TF) μSR measurements. Illumination results in significant enhancements of the depolarization rates of the precession signals by the interaction of photogenerated charge carriers with the three Mu states. A three-state model involving transitions between the Mu states as well as spin-exchange processes gives a good description of the observations [1]. In Ge, light illumination experiments showed that the observed diamagnetic Mu precession signal consists of two diamagnetic states, $\mathrm{Mu_{BC}^+}$ and $\mathrm{Mu_T^-}$, where the 2nd state is strongly influenced by the presence of photoexcited carriers, which is attributed to a fluctuating hyperfine interaction that originates from charge state fluctuations between $\mathrm{Mu_T^-}$ and $\mathrm{Mu_T^0}$ due to hole capture, i.e. $Mu^- + h^+ \longleftrightarrow Mu^0$ [2]. These examples demonstrate the capability of μSR to investigate charge and spin dynamics of impurity states: information which is difficult to obtain by other experimental techniques.

The feasibility of photo-induced experiments on the existing LE-μSR apparatus [10, 11, 12] at the Swiss Muon Source SμS has been tested by the setup shown in Fig. 1. As a light source, commercially available green or white LEDs with a total electrical power of about 3 W are mounted on the radiation shield of the LE-μSR cryostat. The light is generated by 33 InGaN LEDs mounted in series on a ring inside the radiation shield of the sample cryostat. Two types of LEDs have been used: white LEDs (Avago HLMP-CW36-UX00, peak wavelengths at 460 nm and 560 nm) and green LEDs [Kingbright L-7113VGC-Z, peak wavelength 525 nm (2.36 eV), HWHM 39 nm]. The maximum current/voltage rating at room temperature is 30 mA / 3.3 V. The viewing angle $2\Theta_{1/2}$ – where $\Theta_{1/2}$ is the angle with respect to the optical center line where the luminous intensity is 1/2 of the center line value – is 30° and 20°, respectivlely, i.e. the light is emitted in forward direction in a narrow cone. Temperature calibration measurements at the sample position at maximum LED current showed, that the sample temperature increase is < 0.5 K at base temperature (4K), and less at higher temperatures. At 30 mA LED current we measured with a commercial lux meter an intensity of about 10 mW/cm^2 at the sample position. In Si the induced steady state charge carrier concentration n_{eh} at 10 mW/cm^2 of green light (2.36 eV) can be estimated by

$$n_{eh} \approx (1-R)\alpha n_\gamma \tau = 0.65 \cdot 10^4/\mathrm{cm} \cdot 2.6 \times 10^{16} \gamma/(\mathrm{cm^2 s}) \cdot 2\mu\mathrm{s} \simeq 3.5 \times 10^{14}/\mathrm{cm}^3 \qquad (1)$$

with R the reflectivity of Si at 2.36 eV, α the absorption coefficient of Si at 2.36 eV, n_γ the photon flux, and τ the