EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Measuring the sublattice displacement in $Ge_{1-x}Sn_xTe$-based Rashba semiconductors using hyperfine interactions

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Abstract

Electronic phenomena emerging from non-trivial band topology have become a topical subject in condensed matter physics. Several classes of materials have emerged which can be considered newly discovered states of matter, including topological insulators and, more recently, Rashba semiconductors. We propose to investigate the link between crystal symmetry breaking and the Rashba effect in $Ge_{1-x}Sn_xTe$-based Rashba semiconductors using hyperfine techniques. The particularity of the present LOI is that we propose to use long-lived activity during the long shutdown (LS2), either from previously proton-irradiated ISOLDE targets or from material activated externally (cyclotron facility) and loaded into an ISOLDE target.

Introduction

Phenomena emerging from relativistic electrons in solids has become a topical subject in condensed matter physics. The spin-orbit interaction couples the spin degree of freedom with the electronic orbits, therefore providing a link between spin and crystalline structures. For long disregarded as a "weak" interaction, spin-orbit coupling (SOC) has become a rich source for novel physics. Among a wealth of intriguing new physics, several classes of materials and phenomena have emerged which are essentially newly discovered states of matter, including topological insulators and, more recently, Rashba semiconductors. The Rashba effect is a momentum-dependent splitting of the electronic bands in 2-dimenional electron systems (the surface states in the case of Rashba semiconductors). The splitting results from the spin-orbit coupling and an asymmetry in the potential in the direction perpendicular to the 2-dimensional plane. In addition to the fundamental interest in these new quantum states of matter, several fields of application have been proposed, namely spintronics (electric control of the spin) [Bercioux2015, Sante2013] and topological quantum computation (Majorana bound states in p-wave superconductors) [Sato2009].

The larger the symmetry breaking, the larger the Rashba spin splitting becomes. This is exemplified by the giant spin splitting recently discovered for polar bismuth tellurohalide semiconductor compounds BiTeX (X=I, Br, or Cl) [Ishizaka2011, Landolt2012]. However, these materials lack an important property for functionalization, namely, the possibility to switch or tune the spin texture by external means. This limitation can be overcome by materials displaying Rashba-splitting coupled to ferroelectricity: the ferroelectric Rashba semiconductors (FERS) [Picozzi2014, Sante2013, Liebmann2016] where the inversion symmetry is naturally broken by the ferroelectric polarization induced by the anion-cation sublattice displacement (Fig 1 a). As a result, the spin splitting in ferroelectric Rashba semiconductors (FERS) can assume exceedingly high values [Picozzi2014, Liebmann2016].

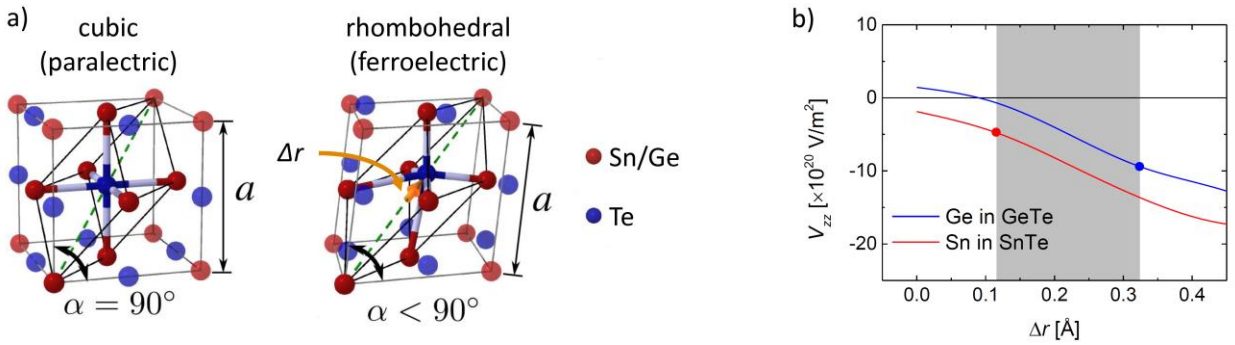

Fig. 1: (a) Lattice structure of $Ge_{1-x}Sn_xTe$ in the cubic (paraelectric) phase and in the rhombohedrally distorted (ferroelectric) phase. The orange arrow indicates the sublattice displacement Δr: the relative displacement between cation (Sn/Ge) and anion (Te) sublattices. (b) z-component of the electric field gradient (V_{zz}) calculated using density functional theory (Wien2k) as a function of Δr. The solid symbols indicate the Δr values for the binary cases (GeTe and SnTe); the grey area indicates the range of Δr for ternary $Ge_{1-x}Sn_xTe$ (calculations underway).

SnTe and GeTe undergo a transition between cubic (paraelectric, above a critical temperature T_C) and rhombohedral (ferroelectric, below T_C) structure. This transition consists of the elongation along the cube diagonal (111) accompanied by a sublattice displacement (relative displacement between Sn/Ge and Te sublattices), and can therefore be described by two parameters: the angle α between **a** and **b** lattice vectors; the sublattice displacement Δr of the atom which would otherwise be in the geometric center of the rhombohedral cell (orange arrow in Fig. 1 a). The magnitude of the distortion and the associated T_C can be tuned by the composition x in $Ge_{1-x}Sn_xTe$ between the binary cases: GeTe with Δr ~0.25 Å and T_C ~670 K; SnTe with Δr ~ 0.07 Å and T_C ~100 K.

Objective

In this letter of intent, we propose to measure the sublattice displacement Δr using hyperfine interaction (HFI) techniques. Complementary studies involve measuring the rhombohedral distortion angle α using synchrotron radiation X-ray diffraction (SR-XRD) and measuring the Rashba splitting using with angle-resolved photoemission spectroscopy (ARPES). Our goal is to experimentally establish the link between symmetry breaking (Δr) and the Rashba effect.

Method

X-ray diffraction, although ideal to measure the lattice constant and α, is not suitable to measure the sublattice displacement [Fons2010, Mitrofanov2014]. The standard technique to probe such local distortions is extended X-ray absorption fine structure (EXAFS). High precision in Δr (ideally ~0.01 Å in the present case) can only be achieved in the near-bulk regime [Fons2010, Mitrofanov2014]. However, both fundamental and applied research on these systems is more generally performed on epitaxial thin films, where the overall lower intensities (and other technical limitations, such as substrate absorption and diffraction artifacts) strongly limit the precision in Δr achievable by EXAFS. Here, we propose to determine the sublattice displacement by measuring the nuclear quadrupole splitting (QS) of selected eMS and PAC isotopes. The QS is given by the product of the nuclear quadrupole moment Q, a property of the nucleus, and the local electric field gradient (EFG) in the neighbourhood of the nucleus. Since the EFG is determined primarily by the electrons involved in the bonding with the nearest neighbours, the QS is extremely sensitive to subtle local distortions, as the rhombohedral distortion described above. By comparing the measured z-component of the electric field gradient (V_{zz}) to density functional calculations of V_{zz} as a function of Δr (Fig. 1 b), we expect to be able to determine Δr with a precision of the order of 0.01 Å. We propose to probe the Sn sublattice using emission Mössbauer spectroscopy (eMS) and perturbed angular correlation (PAC) spectroscopy from radioactive ^{119m}Sn ($t_{1/2}$ = 293 d). For the Ge sublattice, we propose to use PAC spectroscopy from ^{73}Ge by implanting the parent ^{73}As ($t_{1/2}$ = 80 d) isotope. This approach and methodology is closely related to the recently approved experiment IS612 ("Unravelling the local structure of topological crystalline insulators using hyperfine interactions") which also involves studies on $Ge_{1-x}Sn_xTe$. The particularity of the present LOI is that **we propose to use long-lived activity during the long shutdown (LS2)**, **either from (A) previously proton-irradiated ISOLDE targets or (B) from material activated externally (cyclotron facility) and loaded into an ISOLDE target.**

^{119m}Sn can be produced on-line with an UC_x target and a Sn RILIS ion source. Yields of ^{119m}Sn have been measured to $1.8\times10^9/\mu$C, accompanied by about $1.0\times10^9/\mu$C ^{119g}Sn [Koster08]. A standard UC_x target that has been irradiated "cold" for one day with 1 μA proton beam and stored for half a year before reheating and ionizing with the RILIS would allow to implant off-line about 10^{14} ions ^{119m}Sn. ^{73}As is produced at ISOLDE usually with ZrO_2 targets and VADIS ion source with a reference yield of 10^9 ions/μC. If such a target were irradiated "cold" for one day with 1 μA proton beam and stored for 3 months, a total of 4×10^{13} ions ^{73}As could be implanted off-line. Alternatively ^{73}As can be produced at small cyclotrons by Ge(p,n) or Ge(d,n) reactions, followed by As separation by dry distillation from GeO_2 (which can be done in the target unit). Assuming a VADIS efficiency of 10% an initial activity of 40 MBq ^{73}As are required to implant 4×10^{13} ions. This level of activity can be easily handled and the low-energy gamma rays of ^{73}As are easily shielded, already by the 1mm thick Ta target container. The table below gives an estimate of the required number of atoms and associated activity per sample (10 – 20 samples per isotope).

beam	$t_{1/2}$	*N* atoms	fluence (cm^{-2})	Bq
^{119m}Sn	293 d	5×10^{12}	3×10^{12}	1.5×10^5
^{73}As	80 d	1×10^{12}	5×10^{12}	1×10^5

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