Spin dynamics in the topological elemental gray Tin from first principles perspective

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Gray Tin is attracting much more interest as a topological quantum material, which has a precisely controlled composition and can be a material for spintronic devices. However, the spin dynamics in gray Tin is largely unknown. In this paper, we calculate the topological surface state of gray Tin by combining density functional theory and a tight-bind model, also propose an electromagnetics – quantum mechanics hybrid approach on how to enhance with metastructures the spin polarized photocurrent in gray Tin, which was experimentally measured recently.

I. INTRODUCTION

A. Discovery of integer quantum hall effect

In 1980, physicist Von Klitzing measured the Hall resistance in a sample in extremely low temperature and a strong magnetic field, as shown Figure 1. As illustrated in the upper panel in Figure 2, it was found that the Hall resistance shows a “step-wise” with the strength of the external magnetic field and can be written as a combination of fundamental physical constant and an integer, \( \nu \), as shown in Equation 1.

\[
\rho = \frac{2\pi \hbar}{e^2} \frac{1}{\nu}
\]  

(1)

That means the Hall resistance is quantized! The quantum Hall effect is the most important discovery in condensed matter physics in 20th century.

B. Quantized anomalous Hall effect and topological materials

FIG. 1. Illustration of quantum Hall effect experiment.

Since the discovery of quantum Hall effect, extensive theoretical work has been devoted to understanding it. The Hamiltonian of electrons in a magnetic field is as follow,

\[
H = \frac{1}{2} \nu v^2 
\]  

(2)

\[
v = \frac{1}{\mu} [\vec{P} - q \vec{A}(x, y, z)]
\]  

(3)
As shown in above Equations (2), (3), and (4), energy of electrons in an external magnetic field is quantized into discrete energy levels, called Landau levels. Landau quantization is essential to interpreting the quantum aspect of the quantum Hall effect. However, a difficulty arises when attempting to explain the quantum Hall conductivity, simply the inverse of the quantum Hall resistance in Equation (1), which would not have occurred between the gaped Landau levels. Finally, the problem was solved by physicist Thouless et al using ideas from topology. In fact, the Hall conductivity is due to the conductive state quantized with the topological invariant, v, as shown in Equation (5), which turns out to be the same integer as that in Equation (1). Therefore, the conductive state is also called topological state. [2]

\[ \sigma = \frac{e^2}{2\pi h} \nu \]  

(5)

The novel properties of quantum Hall effect raised an important question – can we find the quantum Hall effect in quantum matter in the absence of an external magnetic field? Indeed, the materials having strong spin-orbit coupling could be ideal candidates, and why? As demonstrated theoretically, the spin-orbit coupling plays the role of a magnetic field, which splits electron spin up and down. [3] These materials are therefore known as topological quantum materials. [4] This is a promising beginning! Yet, more important features of topological quantum materials need to be explored. Let us consider the Hamiltonian of a quantum material by including the Rashba spin-orbit term as written below, [5]

\[ H = \frac{k^2}{2m} I + \alpha (\sigma \times k) \cdot \hat{z}, \]  

(6)

where, the Rashba spin-orbit coupling, the second term, conveys some very important information about topological quantum materials. The topological states only exist on the surface of a material, where the inverse symmetry is broken, the Rashba term will vanish otherwise. That says the topological materials are conductive at the surface and insulating in the bulk. Moreover, the spin and momentum are locking. That is, the electron spin perpendicular to its momentum. Electrons with opposite spins move in opposite directions and form a pair of helical surface states, as illustrated in Figure 3.

Topological quantum materials are a promising platform for sensing and quantum information processing. Topological materials supporting helical spin current can detect the helicity of molecules. [6, 7] The numbers in quantum computing can be stored in spin qubits. [8, 9] Driven by the fundamental scientific interests and unprecedented technical applications, discovering new topological quantum materials and exploring their unique properties are the exciting topics. The first 3D topological quantum materials ever discovered are Bi$_{1-x}$Sb$_x$ alloys. [10] Our interest focuses on elemental topological quantum materials, in particular, gray Tin, which is an important semiconductor material can be integrated into devices, whose composition is easily controlled compared with alloys. However, its quantum features have remained largely unknown.

Here, we investigate the topological surface states and \textit{ab - initio} perspective on spin dynamics in gray Tin by a combined theoretical and experimental approach. Section II devotes to the methods. Section III discusses major results, and the key findings are summarized in section IV.

II. METHODOLOGIES

To achieve the desired properties of materials which is governed by the structure-property relationship, well-controlling of a material structure at the atomic level is essential. Therefore, molecular beam epitaxy is an ideal method for this purpose and is chosen to grow gray Tin samples. [11–13]

In order to understand experimental results and to bring a theoretical insight into fundamental physics, the electronic band structure of gray Tin was calculated using the density functional theory using Vienna Ab initio Simulation Package (VASP) program. [15] The exchange-correlation term, $V_{xc}$ was introduced to correct the energy difference between the fictitious non-interacting system and the real system in Equation (6). More importantly, the spin-orbit interaction $\xi(r)L\cdot S$, which plays a crucial role in topological phase, was also included. The topological surface states in $\alpha$-Sn is calculated using the tight-binding model, which uses Wannier function parameters extracted from DFT bulk calculations, efficiently calculating the half-infinite surface with the iterative Green’s functions. [16]
\[ i\hbar \frac{\partial \psi(x)}{\partial t} = -\frac{\hbar^2}{2m} \frac{\partial^2 \psi(x)}{\partial x^2} - \sum_k \frac{Z_k}{|r_i - r_k|} \psi(x) + \int \frac{\rho(r')}{|r_i - r'|} dr' + V_{xc} + \xi(r) \mathbf{L} \cdot \mathbf{S} \] (7)

FIG. 4. (a) Calculated electronic band structure, and (b) projected density of states of gray tin bulk. The optical transition between \( \Gamma_{7c}^- \) to \( \Gamma_{8v}^+ \), \( \bar{E}_0 \), occurs between 4s to 4p orbital.

III. RESULTS

A. Bulk electronic band structure

The good starting point is to investigate the electronic band structure of bulk gray Tin. The electronic band structure of bulk gray Tin demonstrates two notable features: (1) the inverted band structure, where the conduction band is below the valence band, due to the strong spin-orbit coupling;[4] (2) Bulk gray Tin is a zero-band semiconductor. The projected density of state in Figure 4 (b) provides more detail information regarding the individual contribution of orbitals to the density. The inverted band structure near the \( \Gamma \) point, where the conduction band is below the valance band, shows that the band near \( \Gamma_{7c}^- \) is occupied mainly with 4s orbital, while that near \( \Gamma_{8v}^+ \) is dominated with 4p orbital. The optical transition between \( \Gamma_{7c}^- \) to \( \Gamma_{8v}^+ \), \( \bar{E}_0 \), occurs between 4s to 4p orbital.

To be a topological quantum material, gray Tn should have insulating bulk. To do that, we grow gray tin samples on InSb substrates. As shown in Figure 5, a small gap of 30 meV is opened due to the lattice mismatch between gray Tin and the substrate, making the strained gray Tin a promising candidate for a topological quantum material.

B. Topological surface state

To evidence the existence of the topological surface state in gray Tin, the electronic band structure of gray Tin slab under epitaxial strain is calculated. We can see the pair of helical surface states are formed between the inverted valence \( \Gamma_{8c}^+ \) and conduction band \( \Gamma_{7c}^- \), where the Dirac point is below the Fermi level, and are time-reverse protected \((E(-\sigma, -k) = E(\sigma, k))\). The calculated surface states are in good agreement with the experimental measurement.

C. \textit{ab – initio} perspective on spin dynamics

Owing to the spin-momentum locking, spin-polarized photocurrents in topological quantum materials might be readily controlled by varying the polarization of the excitation light. Light irradiation on a sample induces a bulk thermoelectric current due to the Seebeck effect and polarized photocurrents moving in the opposite directions on the surface of the sample due to the optical transition from the bulk valence band to the unoccupied surface state, as shown in Figure 7.[18, 19] The absorption edge of the substrate can contribute to photocurrent as suggested by a recent experiment.[20] The polarized photogalvanic effect makes topological materials attractive materials for next generation of electronic devices. The information about the photo-excited spin dynamics provides an insight into precisely and intentionally control the flow directions of the photocurrents, since efficient spin-current generation is crucial for development of novel spintronic devices.[21]

Okada et al. suggest that the circular photogalvanic current in \( \alpha \)-Sn shows a pronounced peak by tuning the Fermi level \( E_F \) near the Dirac point. As demonstrated by Edens et al., the highest peak of photocurrent in \( \alpha \)-Sn is achieved if the incident photon energy matches the interband transition gap.[20] Recently, Sun et al. demonstrate that the photocurrent response can be magnified by the presence of a meta-nanostructure whose optical...
IV. CONCLUSIONS

In summary, we investigated the topological surface state in gray Tin, an elemental topological quantum material, and also proposed theoretical scheme by combining electromagnetic theory and quantum mechanics approach to study how to enhance the spin polarized photocurrent in gray Tin, which was experimentally measured recently, in the presence of metastructures. Our research finding inspires more future research in construct a complete theoretical and experimental approach to study conversion of charge to spin current. This study will shed light on design highly efficient spintronic devices.

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