Rashba Spin-Orbit Coupling in Colloidal Lead Sulfide Nanosheets

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Abstract
We investigated the Rashba-type spin-orbit coupling in colloidal lead sulfide nanosheets and its dependency to the geometry of the nanosheets by performing circular photogalvanic effect measurements.

1. Introduction

Employing the spin degree of freedom instead of the electrical charge is one of the suggested ideas for the future of data processing due to the low power consumption and the superior functionality of this concept. Colloidal chemistry as an easy and inexpensive method for producing high-quality nanomaterials can be employed to synthesize the required materials for spin-dependent components. In order to evidence that, we have investigated the spin-orbit coupling (SOC) in colloidal lead sulfide nanosheets and its dependency to the geometry of the nanosheets by performing circular photogalvanic effect (CPGE) measurements. Although the PbS rock salt crystal has point symmetry, asymmetric interfaces on top of the material and underneath, as well as the gate electric field can affect the confined crystal and structurally break the symmetry, leading to the Rashba type SOC generation. Our results show that this effect can be tuned by altering the lateral size (width) of the nanosheets. Spin-orbit coupling is a key feature in spintronics which allows manipulating the spin direction by means of external electric fields. The observation of this effect in colloidal materials can be a breakthrough for future inexpensive spintronic devices.

By approaching the end of the Moore's law, it is crucial to develop new concepts or new materials for the future of data processing [1,2]. Using the spin/valley degree of freedom instead of the electrical charge is among the suggested ideas, leading to the introduction of spintronics and valleytronics concepts [3,4]. Up to now, these concepts have been mainly investigated in 2D materials produced by cost-intensive methods such as molecular-beam epitaxy (MBE) [1,5]. Here we show that materials synthesized by colloidal chemistry have the potential to be employed for spintronics, by realization of Rashba spin-orbit interactions through circular-photogalvanic effect measurements. Colloidal synthesis of nanomaterials offers the possibility to inexpensively produce high-quality crystals. Among the colloidal materials, 2D lead sulfide nanosheets show promising properties [6,7]. Although their rock salt crystal structure is centrosymmetric, the symmetry can be broken by the application of asymmetric boundaries underneath and above the crystal (SiO_2 and vacuum) as well as a gate electric field, leading to Rashba type splitting in the band structure. The effect can be tuned by altering the gate voltage, the thickness of the nanosheets (confinement) [1] and their lateral dimensions.

2. Device preparation

Lead sulfide nanosheets have been synthesized following the reported approaches [7] and spin-coated on a Si/SiO_2 substrate. Individual nanosheets were contacted with Ti/Au by means of electron-beam lithography and metal evaporation following by the lift-off process. Then, the sample was transferred to a vacuum probe station and measured at room temperature. In order to observe the circular-photogalvanic effect, a circularly-polarized laser beam (λ=627 nm) was obliquely illuminated to the sample, and the generated current was measured while the bias voltage was kept to zero.

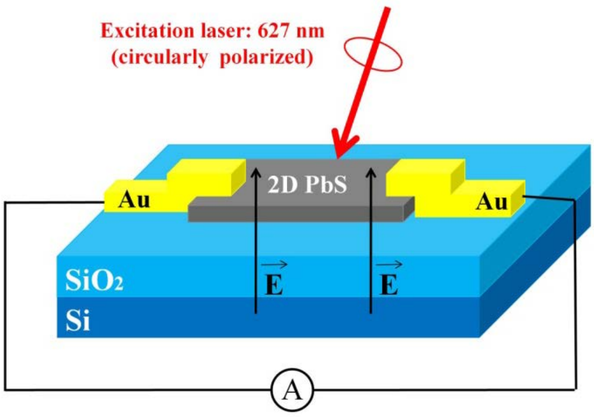

Fig. 1 Schematic of the test setup, which breaks the inversion symmetry by a gate electric field and by asymmetric vertical interfaces (SiO_2 and vacuum).

3. Results and discussion

Figure 1 schematically illustrates the setup of the CPGE measurements. By the application of a gate electric field and asymmetric vertical interfaces, this setup reduces the symmetry in the crystal from the O_h point group (inversion symmetric) to C_{4v} (asymmetric). As we recently showed in [1], breaking the symmetry in the PbS nanocrystal results in splitting the valence and the conduction bands at the M point valleys, corresponding to the (110) direction, at the corners of the rectangular brillouin zone.

The angular momentum of such bands becomes different for the spin-up and spin-down carriers but remains equal for the M and M' valleys (valleys with opposite directions). Therefore, upon illumination with circularly-polarized light, photoexcited carriers are spin polarized but valley un-polarized [1].

Figure 2 shows the photocurrent of the nanosheets at 0 V bias and in different angles of the used quarter-wave plate, corresponding to different helicities of the circularly-polarized light (V_g= -10 V). The measured photocurrent includes a background current originating from the photovoltaic effect (I_0), a contribution from the linear-photogalvanic effect (I_{LPGE}), and eventually, the circular-photogalvanic current (I_{CPGE}), a helicity-dependent current which is attributed to the Rashba SOC in the crystal [1,4,8,9]. Population of the split bands with spin-polarized carriers results in asymmetric distribution of the carriers in momentum space (compared to the Γ point), and generation of I_{CPGE} [1,4,9]. By fitting the measured photocurrent by employing eq. (1) (φ is the angle of the quarter-wave plate), I_0 and I_{CPGE} are obtained equal to 28 and 4.6 pA respectively.

$$J_{\text{total}} = J_0 + J_{\text{CPGE}} \sin(2\varphi) + J_{\text{LPGE}} \sin(2\varphi)\cos(2\varphi) \qquad (1)$$

In order to exclude the spin-independent effects on I_{CPGE}, such as light absorption capability, the normalized CPGE (I_{CPGE}/I_0) is calculated to show the strength of the Rashba effect. As reported in [1] for a 2 µm wide nanosheet with a thickness of 10-12 nm, the normalized CPGE can reach to 7 %. Here, although the thickness is similar to the aforementioned nanosheets, by reducing the lateral size (width=100 nm), this parameter can be improved to 16 %.

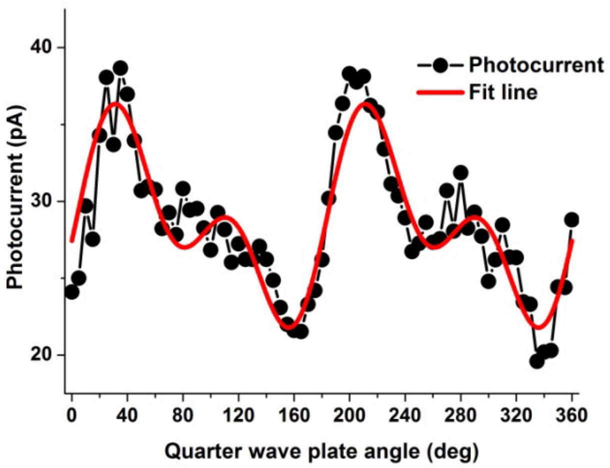

Fig. 2 Variation of the photocurrent at 0 V bias by changing the angle of the quarter-wave plate (light helicity). The CPGE current constitutes 16 % of the photocurrent (V_g = -10 V).

By confining the crystal also in the width, the effect of the back gate originating electric field on the crystal becomes more pronounced. By intensifying the effective electric field, higher degrees of asymmetry are imposed to the crystal which leads to the wider splitting of the bands. Therefore, a higher normalized CPGE current can be detected through illumination of the crystal with circularly-polarized light [1].

4. Conclusions

Rashba spin-orbit coupling has been investigated in colloidal lead sulfide nanosheets with reduced widths. Circular-photogalvanic measurements showed that Rashba band splitting can occur as a result of a gate electric field and asymmetric interfaces to the crystal. The effect could be improved by reducing the width of the crystal. Our observation suggests that colloidal chemistry can support the recent spintronic approaches by inexpensively providing the required active material.

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