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Surface spin polarization in the magnetic response of GeTe Rashba ferroelectric

A. A. Avakyants, N. N. Orlova[1]), A. V. Timonina, N. N. Kolesnikov, E. V. Deviatov

Institute of Solid State Physics of the Russian Academy of Sciences, 142432 Chernogolovka, Russia

I. Introduction. Recent renewal of interest to semimetals is mostly connected with topological effects [1]. Among nonmagnetic topological Weyl semimetals (WSM), GeTe is of special interest [2–4] due to the reported giant Rashba splitting [4–7]. GeTe is predicted to be topological semimetal in ferroelectric α-phase [8, 9]. Nonlinear Hall effect has been demonstrated in GeTe [10], which is the direct manifestation of finite Berry curvature in topological media [11]. The direct measurement of the Rashba-split surface states of α-GeTe(111) has been experimentally realized thanks to K doping [12]. It has been shown that the surface states are not the result of band bending and that they are decoupled from the bulk states. The giant Rashba splitting of the surface states of α-GeTe is largely arising from the inversion symmetry breaking in the bulk [12].

Thus, one can expect a complicated response of a topological semimetal GeTe on the external magnetic field due to the correlation between ferroelectricity and spin textures in GeTe [13], similarly to magnetoelectric structures [14].

GeTe single crystals were grown by physical vapor transport in the evacuated silica ampule. The powder X-ray diffraction analysis confirms single-phase GeTe. To investigate magnetic properties, we use Lake Shore Cryotronics 8604 VSM magnetometer, equipped with nitrogen flow cryostat. A small (0.82–9.54 mg) flake is mounted to the sample holder by low temperature grease, which has been tested to have a small, strictly linear magnetic response. We investigate sample magnetization by standard method of the magnetic field gradual sweeping between two opposite field values to obtain magnetization loops.

All three samples show clear low-field hysteresis in Fig. 1. To our surprise, the saturation level is negative
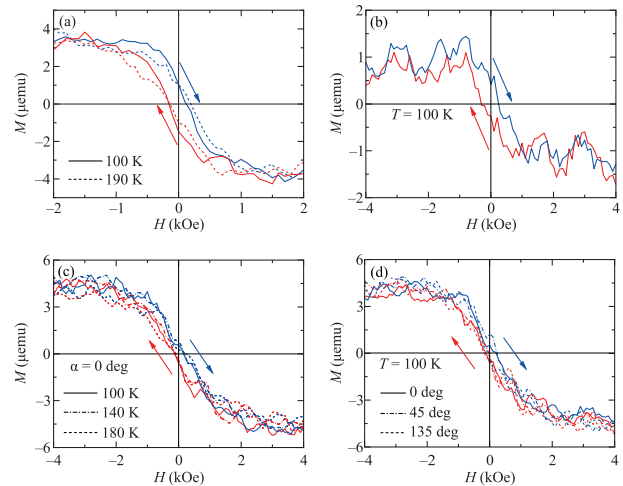
[1])e-mail: honna@issp.ac.ru

Fig. 1. (Color online) The low-field hysteresis region for all three samples. For every field sweep direction, we use curve averaging (8 curves) to increase the signal/noise ratio. The diamagnetic slope is subtracted from the averaged curves to highlight the nonlinear low-field behavior. (a) – Curves for the 6.69 mg GeTe flake at two temperatures, 100 K (solid) and 190 K (dash). (b) – $M(H)$ curves for the smallest, 0.82 mg GeTe flake, at 100 K. (c), (d) – Hysteresis for the 9.54 mg flake at different temperatures (c) and sample orientation (d). For every sample, the saturation level is negative in positive fields, and the loop is passed clockwise, in contrast to usual ferromagnetic hysteresis

in positive fields, and the loop is passed clockwise, in contrast to usual ferromagnetic hysteresis.

We wish to note, that our unusual diamagnetic loop can not be considered as inverted hysteresis in the common sense (e.g. in terms of [15] and references therein). Indeed, usual inverted hysteresis implies two magnetic phases: the inversion reflects the phase interaction in this case, so one magnetic phase provides a bias field to the second one [16]. This bias field forces the magnetization reversal even before the reversal of the external field, so the loop is passed clockwise. In our case, we observe unusual diamagnetic loop, where the satu-

ration level is inverted instead of the switching field. The saturation level is negative in positive fields, it is reversed after the external field reversal. Also, the remanence plots technique (i.e., Henkel or δM plots) does not confirm several magnetic phases for our GeTe flakes.

The experimental curves in Fig. 1 can not be continuously transformed to the standard ferromagnetic one by adding/subtracting of any linear dependence, which excludes any possible contribution from any magnetic contamination, e.g. magnetic impurities [17]. To rule out systematic error of the VSM, like possible remanent field in the electric magnet, etc., we demonstrate strictly linear diamagnetic dependence without GeTe sample for the same setup, the same sample holder, the same grease, and at the same temperature.

For these reasons, we should consider possible contribution from the surface-state induced spin textures [12] in α-GeTe(111). Direct correlation between ferroelectricity and spin textures was demonstrated in GeTe [13]. Both the giant Rashba splitting of the surface states and bulk ferroelectricity are largely arising from the inversion symmetry breaking [12]. Thus, GeTe single crystal can be considered as magnetoelectric heterostructure [14].

In the conditions of our experiment, variation of the magnetic field leads to appearance of the electric field due to the magnetoelectric coupling [14, 18]. Electric field affects spin textures in GeTe [13], which, subsequently, affects magnetization response. In this case, the unusual diamagnetic loop appears due to the delay of the magnetic response to the external field, since electric fields are coupled to strain in ferroelectrics [14]. Thus, the unusual diamagnetic loop is a direct consequence of correlation between ferroelectricity and spin-polarized surface states in GeTe.

We wish to thank S. S. Khasanov for X-ray sample characterization.

Conflict of interest. The authors of this work declare that they have no conflicts of interest.

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