Semiconductors, 2023, Vol. 57, No. 4

Peculiarities of surface structure and surface electron transport in correlated topological insulator SmB_6

© *E.A. Artemov*[1,2], *A.V. Mantuzov*[1,2], *V.S. Zhurkin*[1], *A.D. Bozhko*[1], *O.S. Kudryavtsev*[1], *B.V. Andryushechkin*[1], *V.M. Shevlyuga*[1], *N.Yu. Shitsevalova*[3], *V.B. Filipov*[3], *V.V. Glushkov*[1]

[1] Prokhorov General Physics Institute of the Russian Academy of Sciences,
119991 Moscow, Russia
[2] AEM Technologies LLC,
109651 Moscow, Russia
[3] Frantsevich Institute for Problems of Materials Science National Academy of Sciences of Ukraine,
03142 Kyiv, Ukraine

E-mail: artpoliofan@mail.ru

Received May 5, 2023
Revised June 29, 2023
Accepted July 6, 2023

New method of chemical-mechanical polishing (CMP) with compositions based on nanometer-sized amorphous silica particles has been developed for the treatment of the surface of single crystals of samarium hexaboride SmB_6. It is shown that the CMP method makes it possible to achieve surface roughness of the SmB_6 single crystals for a defect-free area with a root-mean-square profile deviation not exceeding 0.8 nm. The effect of the CMP method on the structural and electronic properties of the (100) and (110) surfaces of single-crystal SmB_6 samples is discussed.

Keywords: samarium hexaboride, chemical-mechanical polishing, surface conductivity, topological insulator.

DOI: 10.61011/SC.2023.04.56418.02k

1. Introduction

Samarium hexaboride SmB_6 is distinguished among other correlated topological insulators by an extraordinary sensitivity of parameters of surface electron transport to the method of preparation of single crystal faces [1–3]. A reliable procedure of preparation of a quality surface of SmB_6 single crystals [4] is a prerequisite for systematic and reproducible studies of the surface defects of SmB_6 and their influence on the parameters of two-dimensional Dirac charge carriers. Mechanical and (or) chemical polishing (etching) is used most often at present for SmB_6 surface preparation. Suspensions based on diamond powders, Al_2O_3, or SiC are used for mechanical polishing of surfaces [5,6]. However, the structural perfection of a near-surface layer, which may vary in thickness from several tens of nanometers to tens of micrometers, is violated essentially in the course of surface processing with such suspensions [7]. It is also known that polished samarium hexaboride surfaces subjected to chemical etching (e.g., with an aqueous solution of nitric acid) show a reduction in the Hall concentration of surface carriers (by more than two orders of magnitude) and an increase in resistivity (by an order of magnitude) [2,3]. It has been demonstrated in [3] that the effective parameters of surface conductivity depend both on the method of special surface treatment and on the surface polarity. The etching of polished polar SmB_6 surfaces formed by (100) planes initiates a reduction in concentration and an increase in mobility of surface conduction electrons at 1.9 K from $113/a^2$ (lattice parameter $a \approx 4.134$ Å) and $1.12\,cm^2/(V \cdot s)$ to $0.76/a^2$ and $18\,cm^2/(V \cdot s)$, respectively [3]. However, chemical etching does not just leave the roughness of mechanically polished surfaces formed by lattice planes (100), (110), and (111) unchanged; instead, it enhances considerably (by a factor of 2–4) the roughness of surface relief [3].

The quality of SmB_6 surface preparation achieved after mechanical polishing or chemical etching [3] is too low to apply scanning tunneling microscopy (STM) techniques to examine SmB_6 surface defects and their influence on the parameters of two-dimensional Dirac carriers. Thus, the issue of preparation of surfaces with different orientations (formed by lattice planes (100), (110), or (111)) with roughness at the level of 1 nm for experimental samarium hexaboride samples is highly relevant. The aim of the present study is to develop a conceptually novel method of surface preparation for SmB_6 single crystals based on a versatile, reproducible, and highly efficient chemical-mechanical polishing (CMP) technique [8] and examine the structural and transport properties of surfaces prepared this way.

2. Experimental procedure

Samarium hexaboride single crystals were grown by crucible-less induction zone melting with double passage of the zone in argon atmosphere. Plates with surfaces oriented along lattice planes (100) and (110) were cut from the grown cylindrical single crystals. The thickness and transverse dimensions of single-crystal plates were 0.675–0.7 and 6–8 mm, respectively. Preliminary processing of the surface of single-crystal SmB_6 was performed by polishing with diamond powders, and finishing CMP treatment was carried out with the use of table-top „Presi“ units. A Bruker D8 Discover A25 X-ray diffractometer was used to control the accuracy of orientation of the sample surface and verify that the surface was non-mosaic. The deviation of orientation of sample faces from the related lattice planes was $\leq 1-2°$.

In order to examine the influence of CMP on the samarium hexaboride surface properties, CMP-treated surfaces were subjected, following a series of measurements of the transport properties, to polishing with diamond powder with a grain size up to $0.3\,\mu$m and subsequent etching in an aqueous solution of HNO_3 $(1:2)$ performed for 5 min.

Measurements of the microrelief of polished and etched surfaces were carried out using an NT-MDT NTEGRA Spectra atomic force microscope and a GPI-300 scanning tunneling microscope. The resistivity and the Hall effect were examined in consecutive experiments in the four-point geometry with linear (for the resistivity) and transverse (for the Hall effect) positioning of potential contacts. Current and potential contacts were positioned in the central part of the studied surface at a distance no less than 1 mm from the sample edges in order to reduced the influence of peripheral regions of single-crystal plates. The typical distances between current contacts were 3 mm, and the distances between potential contacts varied from 1 mm (for the resistivity) to 3 mm (for the Hall effect). The DC current amplitude was adjusted within the range from $3\,\mu$A to 10 mA to exclude the possibility of sample overheating. Resistivity measurements were performed at temperatures of 1.9–300 K. The magnetoresistance and the Hall effect were examined at temperatures of 1.9–4.2 K in magnetic fields up to 8 T.

3. Results and discussion

3.1. Surface preparation by CMP

The preparation of SmB_6 sample surfaces by CMP was performed in two stages. At the first stage, planes of single-crystal SmB_6 plates were processed successively with diamond powders ASM 3/2 and ASM 1/0. At the second stage, samples were polished in acidic compositions with amorphous particles of colloidal silicon dioxide used as the solid phase. The particle size varied from 10 to 100 nm.

The surface of SmB_6 samples was polished in the following way. A single-crystal SmB_6 plate was secured to a special mounting setup and mounted on a polishing table with a polishing pad glued to it. CMP processing was performed in different regimes with continuous feeding of the polishing composition. On completion of the CMP process, the polished SmB_6 sample surface was rinsed with warm distilled water, the sample was removed from the mounting setup, the residue of polishing composition and contaminants were removed from surface with a cleaning solution, and the sample was dried.

3.2. Surface structure

Atomic force microscopy (AFM) data revealed that the roughness of surfaces of single-crystal SmB_6 samples prepared by CMP was, in contrast to abrasive polishing and chemical etching [3], independent of their orientation. The obtained surfaces were characterized by low roughness and insignificant curvature (deviation from planarity) of a smooth relief with a characteristic level difference ≤ 20 nm over a length of $\sim 30\,\mu$m.

STM data did also indicate that the surface of single crystals was very smooth on macroscopic scales (Fig. 1, *a*) and that the root-mean-square profile deviation within a typical defect-free section of the surface of SmB_6 single crystals did not exceed 0.8 nm (Fig. 1, *b*). Irregular terraces for (100) planes with various dimensions separated by single- and diatomic steps were identified in the STM images of the SmB_6 single crystal surface (Figs. 1, *c, d*). The high degree of smoothness of surfaces prepared by CMP confirms that this method has a signilicant application potential in studies of the surface structure and the surface electron transport in samarium hexaboride.

3.3. Resistivity

The temperature dependences of resistivity of single-crystal samarium hexaboride samples reduced to the resistivity value at 290 K are presented in Fig. 2. As was expected, the surface preparation exerts no noticeable influence on the resistivity of samples in the region of temperatures corresponding to bulk charge transport ($T > 9$ K). Slight differences in the behavior of $\rho(T)$ below 15 K (Fig. 2) are attributable to the differences in growth conditions of the initial single crystals. It is important to note that the activation energy values corresponding to activation growth of the resistivity in the 9–15 K temperature interval are almost indistinguishable within the limit of experimental error and equal to $\Delta = 56 \pm 1$ K. In the surface conductivity region ($T < 4.2$ K), the resistivity of the studied SmB_6 samples is significantly higher than the corresponding values for a reference single crystal with (100) faces subjected to chemical etching in an aqueous solution of nitric acid (see [3]). The resistivity of plates with (110) surfaces increases with decreasing temperature in accordance with a power law $\rho \sim T^{\alpha}$ with exponents $\alpha \approx -0.57$ and $\alpha \approx -0.38$ for surfaces prepared by CMP (denoted by C) and using the „traditional“ method of abrasive polishing and chemical etching (denoted by E), respectively.

The CMP effect is more pronounced in the case of nonpolar SmB_6 surfaces. The ratio of resistances for (100) surfaces subjected to CMP treatment and „traditional“ abrasive polishing and chemical etching does not exceed 3% at 1.9 K. The corresponding ratio for (110) surfaces is as high as 15% (see the inset in Fig. 2). A noticeable rise of the resistance ratio at lower temperatures in the case of (110) surfaces subjected to CMP and the „traditional“ treatment

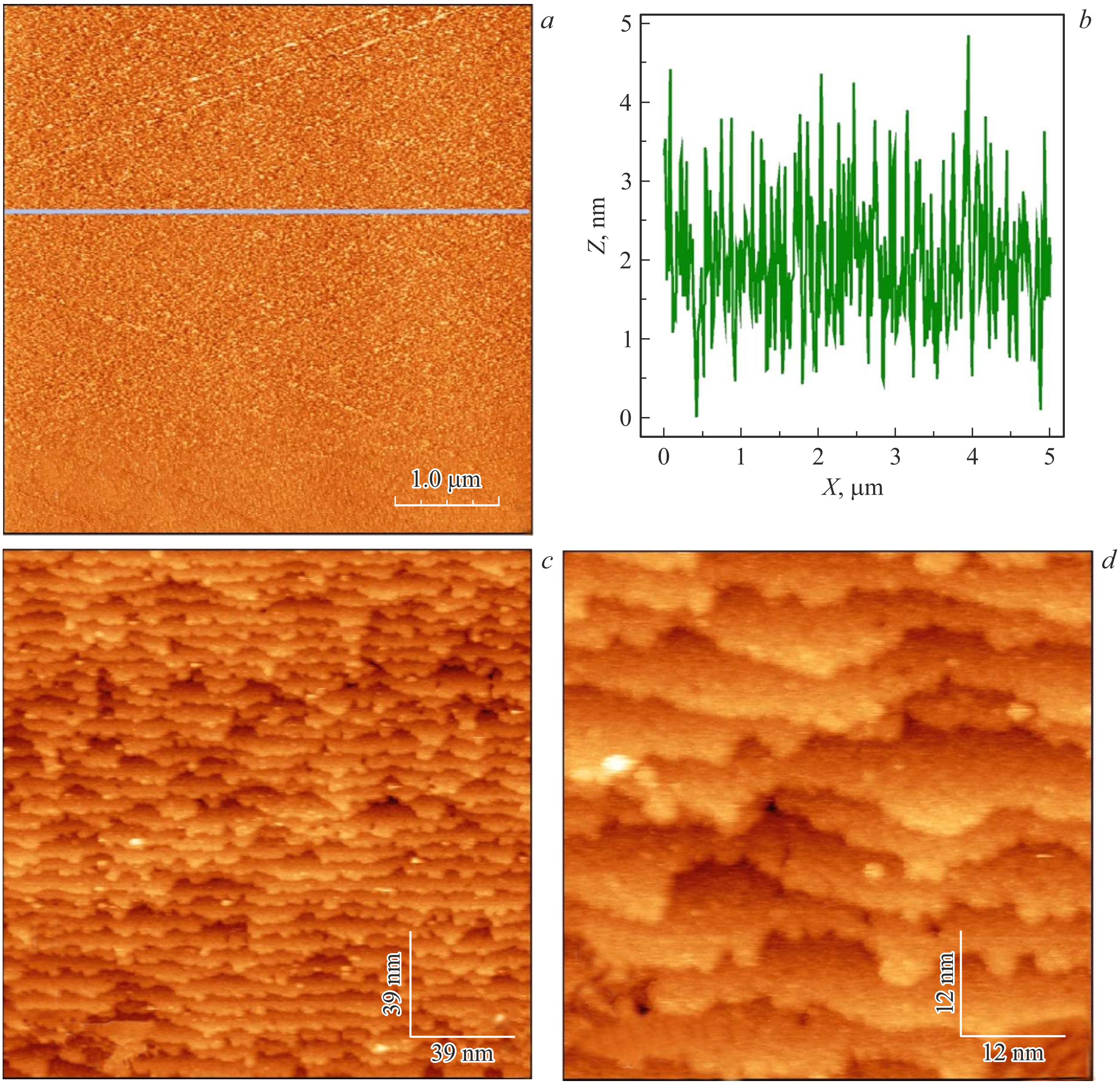

Figure 1. STM images ($I_t = 2\,\text{nA}$, $U_S = -150\,\text{mV}$) of the surface of a SmB_6 single crystal prepared by CMP (dimensions: *a* — $5.2 \times 5.2\,\mu\text{m}^2$, *c* — $194 \times 194\,\text{nm}^2$, and *d* — $46.2{-}49.7\,\text{nm}^2$). Terraces in the lower panels correspond to (100) planes separated by single- and diatomic steps. The surface relief along the line indicated in panel *a* is shown in panel *b* on a fine scale.

should apparently be associated with a reduction in the exponent, which changes from $\alpha \approx -0.57$ to $\alpha \approx -0.38$, after etching (Fig. 2). Note that a lack of a essential power-law resistivity behavior in the sample with the [100] surface (Fig. 2) precludes us from attributing the $\rho \sim T^{\alpha}$ dependences to effects associated with the influence of interelectron interaction in systems with a strong disorder [9], which is induced in SmB_6 by the presence of steps of various dimensions and orientations on the surface (Fig. 1). At the same time, power-law temperature dependences of resistivity are not typical of electron transport in topological insulators, where the processes of carrier scattering are blocked by a rigid coupling between the spin and the momentum of an electron.

3.4. Galvanomagnetic properties

The most significant differences in magnetoresistance were observed in the SmB_6 sample with (110) surfaces at a temperature of 1.9 K. In contrast to (100) surfaces, where the preparation method has almost no influence on the $\Delta\rho/\rho$ magnitude (Fig. 3), CMP of the (110) surface enhances the magnitude of negative magnetoresistance significantly compared to the surface subjected to abrasive polishing and chemical etching (from $\Delta\rho/\rho \approx -13\%$ to $\Delta\rho/\rho \approx 18\%$ in a 8 T field at a temperature of 1.9 K; see Fig. 3). Particularly notable is the field dependence, which is close to linear $\Delta\rho/\rho \sim B$ and may be indicative of electron transport in the regime of a strong disorder [10].

Hall effect data in SmB_6 are shown in the inset of Fig. 3. For ease of comparison, the measured values were normalized to the corresponding value at 4.2 K. It is evident that the growth of the Hall coefficient magnitude for polar (100) surfaces at lower temperatures is comparable to the variation for the reference sample from [3]. At the same time, the Hall coefficient for nonpolar (110) surfaces increases by a factor smaller than 2, and the relative variation is almost independent of the surface preparation method. With high values of resistivity for (110) surfaces (Fig. 2) taken into account, this behavior of the Hall coefficient may be attributed to the low mobility of surface carriers due to scattering by defects and relief irregularities. Estimates of the Hall mobility of surface carriers for nonpolar (110) surfaces vary from 3 to 5.5 $cm^2/(V^{-1} \cdot s^{-1})$, agreeing closely with the known data for surfaces of SmB_6 single crystals [3].

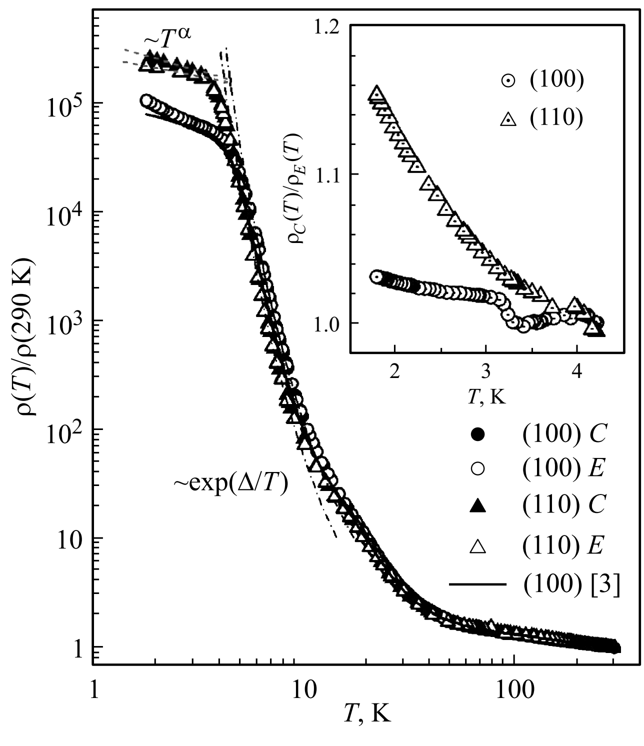

Figure 2. Normalized temperature dependences of resistivity $\rho(T)/\rho(290\,K)$ for single-crystal SmB_6 plates with (100) and (110) surfaces prepared by CMP (C) and abrasive polishing with subsequent chemical etching (E). The solid curve represents the $\rho(T)/\rho(290\,K)$ values for a single-crystal SmB_6 sample with (100) surfaces after etching [3]. Dash-dotted and dashed curves correspond to the activation and power asymptotics of resistivity (see text). The $\rho_C(T)/\rho_E(T)$ ratio for SmB_6 samples with surfaces (100) and (110) within the temperature range corresponding to the regime of surface conductivity is shown in the inset.

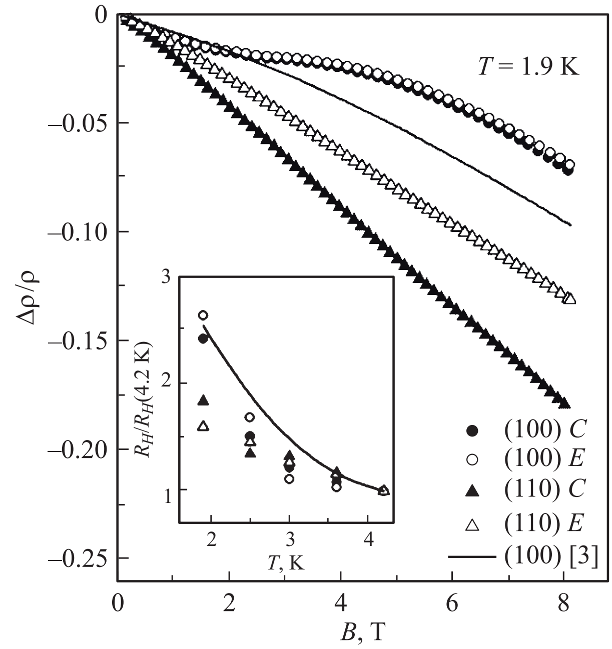

Figure 3. Magnetoresistance $\Delta\rho/\rho(B)$ at a temperature of 1.9 K for single-crystal SmB_6 plates with (100) and (110) surfaces prepared by CMP (C) and abrasive polishing with subsequent etching (E). The solid curve represents the $\Delta\rho/\rho(B)$ values at 1.9 K for a single-crystal SmB_6 sample with (100) surfaces after etching [3]. Ratio $R_H(T)/R_H(4.2\,K)$ for SmB_6 samples with (100) and (110) surfaces prepared using methods C and E is shown in the inset, where the data for a single-crystal SmB_6 sample with (100) surfaces after etching [3] are also presented for comparison.

4. Conclusion

It has been demonstrated for the first time that the application of acidic compositions with amorphous colloidal silicon dioxide nanoparticles serving as the solid phase for CMP of SmB_6 single crystal surfaces oriented along different (polar (100) and nonpolar (110)) lattice planes establishes the conditions for uniform autolysis (polishing) of the material surface. It has been found that CMP is the most efficient when it proceeds via localized electrochemical partial reactions within defect-free regions lying between growth macrodefects; notably, the mean surface roughness of the prepared surface is then characterized by a root-mean-square profile deviation below 0.8 nm. The application of CMP to SmB_6 provides an opportunity to modify significantly the parameters of electron transport for nonpolar surfaces (110). A high degree of surface smoothness with a relatively insignificant relief curvature (up to 20 nm over a length of 30 μm) and the observation of atomic terraces in STM images confirm that this method has a significant application potential for preparation of SmB_6 surfaces with subsequent STM studies of the structural and electronic properties and the examination of effective parameters of surface carriers with the use of the field effect.

Acknowledgments

This study was supported by grant No. 22-22-00990 (https://rscf.ru/project/22-22-00990/) from the Russian Science Foundation. Equipment provided by the „Analytical Center of the Prokhorov General Physics Institute of the Russian Academy of Sciences“ and the Center for Collective Use of the University of Chemical Technology of Russia was used in the study.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by D.Safin