Effects of Spark Plasma Sintering Conditions on the Anisotropic Thermoelectric Properties of Bismuth Antimony Telluride

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1. Supplement SEM images

Fig. S1 SEM of the Commercial Bi0.4Sb1.6Te3 powder

Fig. S2 SEM-EDS element mapping of the specimen sintered by SPS at 653K.
Fig. S3 SEM-EDS element mapping of the specimen sintered by SPS at 773K.
2. SPS sample cutting schematics.

![SPS sample cutting schematics](image1.png)

**Fig. S4** SPS sample cutting schematics. The red specimens were measured for in-plane thermoelectric properties, and the grey specimens were measured for out-of-plane thermoelectric properties.

3. Heat capacity $C_p$

![Heat capacity graph](image2.png)

**Fig S5.** Temperature dependence of the specific heat at constant pressure for Bi0.4Sb1.6Te3 samples, the red line represents the measured value by DSC, and the dotted line represents the calculated values by Dulong-petit law.
4. Metal droplets squeezed out of graphite die after SPS sintering

Fig. S6 Spilled droplets squeezed out of graphite die after SPS sintering at 723 K (left) and 773 K (right).

5. Calculations of Lorenz number \( L_o \)

According to the measured \( n \) and estimated \( m^* \) values, a simple parabolic band model can be applied by employing the following equations:

\[
F_\lambda(\xi) = \int_0^\infty \frac{x^2 dx}{1 + \exp(x - \xi)} \tag{S1}
\]

\[
L_o = \left( \frac{k_B^2}{q^2} \right) \left( 3F_0F_2 - 4F_1^2 \right) / F_0^2 \tag{S2}
\]

where \( F_\lambda(\xi) \) is the Fermi integral and \( \xi \) is the reduced electrochemical potential. \( \lambda \) is a scattering parameter and 0 is assumed for acoustic phonon scattering, 1 for optical phonons scattering, and 2 for ionized impurity scattering. \( k_B \) is the Boltzmann constant, \( q \) is the unit charge of electron. \( \xi \) is calculated from the measurement Seebeck coefficient (S) using the following equation:
\[ S = -\frac{k_B}{q} \left[ \frac{(2 + \lambda) F_{\lambda + 1}}{(1 + \lambda) F_{\lambda}} - \xi \right] \]
6. Sintering profiles of samples sintered with variable holding time, pressure, and ramp-rate.

**Fig. S7.** (a) Sintering profile for SPS sintering with variable holding time. (b) Mass density of the samples as a function of holding time. (c) Degree of orientation as a function of holding time.

**Fig. S8** (a) Sintering profile for SPS sintering with variable pressure. (b) Mass density of the SPS sintered samples with variable uniaxial pressure. (c) Degree of orientation as a function of uniaxial pressure.
Fig. S9 (a) Sintering profile for SPS sintering with variable ramp-rate. The inset shows the current profile during SPS sintering. (b) Mass density of the samples sintered with variable ramp-rate. (c) Degree of orientation of the samples sintered with variable ramp-rate.