Low temperature thermoelectric material BiSb with magneto-thermoelectric effects

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Thermoelectric (TE) materials

The thermoelectric effect refers to phenomena by which either a temperature difference creates an electric potential or an electric potential creates a temperature difference. [1]

\[
ZT = \frac{\sigma S^2 T}{\lambda}
\]

Bismuth antimony alloy, which was known to be the first topological insulator, was reported to have a significantly higher figure of merit ZT at low temperature range (20K~220K) than the regular room temperature TE materials (ex. bismuth telluride).
Low temperature TE material BiSb

- **Bi85Sb15**
  - Mel-spin+SPS by Luo(2013) [3]

- **Bi0.6Sb1.4Te3** melt-spin nano-structure
  - by W. Xie et al. (2012) [4]

- **Yb0.9La0.1Cu2Si2**
  - by G. J. Lehr et al. (2014) [5]

- **YbAgCu4** ball milled nano-structure
  - by M. Koirala et al. (2014) [6]
Low temperature TE material BiSb

Phase diagram of Bi-Sb solid solution, Cs/CL represents the segregation coefficient.\[2\]

Energy band configuration of Bi1-xSbx alloys as a function of x, at T~0K.
Bismuth: L-electrons, T-holes
Antimony: L-electrons, H-holes
BiSb ZT enhancement in magnetic field

Bi85Sb15 Single Crystal Z33
Yim&Amith (1972) [7]
Magneto-thermoelectric effects

Basic effects in a magnetic field:
Assuming magnetic field $B$ is perpendicular to this plane, outward.
Carriers have negative net charges.

**Hall:**
$j + B \rightarrow E$
$Rh = E/(jB)$

**Ettingshausen:**
$j + B \rightarrow \frac{dT}{dy}$
$P = \left(\frac{dT}{dy}\right)/(jB)$

**Nernst:**
$w + B \rightarrow E$
$N = E/(BdT/dx)$

**Righi-Leduc:**
$w + B \rightarrow \frac{dT}{dy}$
$L = \left(\frac{dT}{dy}\right)/(BdT/dx)$

Rh, P, N, L Correspond to four coefficients

$j$: current flow
$w$: heat flow
$E$: Electric field
$\frac{dT}{dy}$: Temperature gradient

$\text{Rh}, P, N, L$ Correspond to four coefficients
Magneto-thermoelectric effects

Wolfe and Smith(1962)[8] claimed that magneto-Seebeck effects of Bi-Sb alloys are the Hall effect acting on the Nernst effect and, to a lesser extent, the Nernst effect acting on the Righi-Leduc effect, which are called the transverse-transverse thermo-galvanomagnetic effects.

So there are two separate effects gourp:
(a) Nernst effect + Hall effect
(b) Righi-Leduc effect + Nernst effect

Where (a) plays a more important role here.

\[(S^a - S^i)_{jj} = B^2 (N_{jk} R_{kj} + L_{jk} N_{kj})\]

Where \(S\) is thermal power, note a&i correspond to adiabatic and isothermo. \(N\) is Nernst coefficient, \(R\) is Hall coefficient, \(L\) is Righi-Leduc coefficient. Subscripts j&k refer to the directions of the primary flow and the transverse effect.
Nernst effects

Using conductivity tensors, we can express the electrical and thermal currents [9]:

\[
\begin{align*}
\bar{J}_e &= \bar{\sigma} \cdot \bar{E} - \bar{\alpha} \cdot \nabla T \\
\bar{J}_q &= T \bar{\alpha} \cdot \bar{E} - \bar{\kappa} \cdot \nabla T
\end{align*}
\]

Where \(\alpha, \sigma, k\) are the thermoelectric, electric, thermal conductivity tensors, respectively. \(E\) is the electric field.

The solution of Boltzmann equation leads to following link between these two tensors:

\[
\bar{\sigma} = -\frac{\pi^2}{3} \frac{k_B^2 T}{e^2} \frac{\partial \sigma}{\partial \epsilon} \bigg|_{\epsilon = \epsilon_F}
\]

And then with the Hall angle \(\tan \theta = \frac{\sigma_{xy}}{\sigma_{xx}}\) gives:

\[
N = -\frac{\pi^2}{3} \frac{k_B^2 T}{eB} \frac{\partial \tan \theta_H}{\partial \epsilon} \bigg|_{\epsilon = \epsilon_F}
\]

In an energy-dependent first order approximation, we can replace \(\frac{\partial \tan \theta_H}{\partial \epsilon} \bigg|_{\epsilon = \epsilon_F}\) with \(\tan \theta_H\), then use a carrier mobility to substitute Hall angles:

\[
\frac{\tan \theta_H}{B} = \mu = \frac{e \tau}{m^*}
\]

Then we can express the Nernst coefficient \(N\) as:

\[
N = \frac{\pi^2}{3} \frac{k_B^2 T}{e} \frac{\mu}{\epsilon_F}
\]

Which is proportional to the ratio of carrier mobility and Fermi energy.
Magneto-resistance

As a simple approach, based on Drude’s model:
\[
\frac{d}{dt} \vec{p}(t) = q \left( \vec{E} + \frac{\vec{p}(t) \times \vec{B}}{m} \right) - \frac{\vec{p}(t)}{\tau}
\]

Geometrical magnetoresistance:
\[
\rho_B = \frac{E_x}{j_x} = \rho_0 (1 + (\mu B)^2)
\]

Assuming Intrinsic, electrons mobility dominated.

The fields bent the current and forced it to travel through a longer path.
Calculation of Thermal power enhancement in Magnetic field

Using the equation we derived before:

$$(S^a - S^i)_{jj} = B^2 (N_{jk} R_{kj} \sigma_{kk} + L_{jk} N_{kj})$$

Because $R, \sigma >> L$, so the first term plays the major role in this enhancement, and for our polycrystalline sample, we should have isotropic parameters. Then we can get the enhancement on thermal power:

$$\Delta S(B) \approx R_{H} \cdot N \cdot \sigma \cdot B^2$$

$$= -\frac{1}{ne} \frac{\pi^2 k_B^2 T}{3} \frac{\mu}{e \varepsilon_F} \frac{\sigma_0 B^2}{1 + \lambda (\mu B)^2}$$

$$= -\frac{\pi^2 k_B^2 T \mu \sigma_0}{3ne^2 \varepsilon_F} \frac{B^2}{1 + \lambda (\mu B)^2}$$

Data from our Bi85Sb15 sample measurements in different fields

Calculated by the theory model
Material synthesis and Sample measurements

- Furnace melting with L-N2 quenching
- Mechanical alloying -- Ball milling
- Melt-spinning
- Spark plasma sintering (SPS)

- Physical Property Measurement System (PPMS)
- X-Ray Diffraction (XRD) analysis
- Scanning electron microscope (SEM)
- Energy Dispersive Spectroscopy (EDS)
References


Thank you!