Anisotropic Thermopower in Al-Si-Multilayers

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Abstract

Semiconductor-metal multilayer structures A/B/A... show, according to model calculations, large anisotropy in their electrical and thermal transport properties: For aluminum (A) and n-type silicon (B), the anisotropy of the thermopower ΔS = S_{\parallel} - S_{\perp} where S_{\parallel} and S_{\perp} represent the Seebeck coefficients along and perpendicular to the layers, is more than an order of magnitude larger than that for a pure metallic structure A/B/A... made of constantan (A) and copper (B). In samples prepared with a tilt angle between sample surface and layer planes, radiation heating of the sample surface induces a surface perpendicular temperature gradient and, due to the anisotropy of the thermopower, generates a surface parallel thermoelectric field $E \sim \Delta S$. This transverse Seebeck effect suggests applications for radiation detection. The preparation of Al-Si multilayer samples is described and their use as rugged, self-powered and nearly wavelength independent detectors for laser radiation is demonstrated.

Introduction

Anisotropic thermopower recently was observed in the high-temperature superconductors $YBa_2Cu_3O_{7-\delta}$ (YBCO) and $Bi_2Sr_2CaCu_2O_8$ (BSCCO) in the normal conducting state [1,2]. Due to anisotropy, in off-c-axis grown YBCO and BSCCO thin films a surface perpendicular temperature gradient induced by laser heating generates a surface parallel thermoelectric field component giving rise to a transverse thermoelectric response. For thin YBCO films sub-ns response times were observed [3]. Later on, also in tilted Copper-Constantan metallic multilayer structures a transverse thermoelectric response was found [4,5].

For applications based on anisotropic thermopower a large thermoelectric anisotropy ΔS is crucial. A multilayer stack A/B/A... (Fig. 1a) consisting of materials A, B represents an anisotropic medium characterised by transport properties S_{\parallel} , σ_{\parallel} , λ_{\parallel} parallel to the layers and S_{\perp} , σ_{\perp} , λ_{\perp} in perpendicular. Here S, σ and λ represent the absolute thermopower and the electrical and thermal conductivities.



Fig. 1: a) Multilayer stack with transport properties parallel and perpendicular to the layer planes b) tilted multilayer sample prepared from the stack with tilt angle α

Theory

From Kirchhoff's rules we obtain for the transport properties parallel and perpendicular to the layers [5,6]:

$$S_{\parallel} = \frac{S_A \sigma_A + p S_B \sigma_B}{\sigma_A + p \sigma_B} \quad S_{\perp} = \frac{S_A \lambda_B + p S_B \lambda_A}{p \lambda_A + \lambda_B}$$

$$\lambda_{\parallel} = \frac{\lambda_A + p \lambda_B}{1 + p} \qquad \lambda_{\perp} = \frac{\lambda_A \lambda_B (1 + p)}{p \lambda_A + \lambda_B}$$
(1)

where $\sigma_{A,B}$ and $\lambda_{A,B}$ are the electrical and thermal conductivities, respectively, $S_{A,B}$ is the Seebeck-coefficient of materials A and B, and $p = d_B / d_A$ is the thickness ratio of layers B and A with thickness d_B and d_A . According to the tensorial description of the Seebeck effect, the thermoelectric field E due to a temperature gradient ∇T is:

$$\vec{E} = S \ \vec{\nabla} T \tag{2}$$

where S is the Seebeck tensor. For tetragonal symmetry and with the coordinate system of Fig. 1b S is [7]:

$$\mathbf{S} = \begin{pmatrix} S_{\parallel} \cos^2 \alpha + S_{\perp} \sin^2 \alpha & 0 & \frac{1}{2} (S_{\parallel} - S_{\perp}) \sin(2\alpha) \\ 0 & S_{\parallel} & 0 \\ \frac{1}{2} (S_{\parallel} - S_{\perp}) \sin(2\alpha) & 0 & S_{\parallel} \sin^2 \alpha + S_{\perp} \cos^2 \alpha \end{pmatrix}$$
(3)

Therefore, a temperarure gradient $V_z T$ applied in zdirection (Fig. 1b) generates, in a multilayer structure tilted by an angle α , a thermoelectric field component in x-direction:

$$E_x = \frac{1}{2} (S_{\parallel} - S_{\perp}) \cdot \sin(2\alpha) \cdot \nabla_z T \tag{4}$$

giving rise to a voltaic response $U_x = l \cdot E_x$ at sample contacts of a sample with heated length l. Absorption of continous optical power P_{abs} within the sample and heat flow into the sample mount generates, according to the equation of heat conduction, a temperature gradient:

$$\nabla_z T = \frac{P_{abs}}{\lambda_z \cdot F} \tag{5}$$

where $F = l \cdot b$ is the heated sample area, b is the heated sample width, and $\lambda_z = \lambda_{\parallel} \sin(\alpha)^2 + \lambda_{\perp} \cos(\alpha)^2$ is the heat conductivity in z-direction. Therefore,

$$U_{x} = \frac{P_{abs}}{2b} \cdot \frac{(S_{\parallel} - S_{\perp})}{\lambda_{z}} \cdot \sin(2\alpha)$$
(6)

By inspection of eq. 1, it becomes obvious, how to choose materials A and B to obtain large thermoelectric anisotropy: Using a metal (A) and a semiconductor (B) should result in large ΔS , approaching $\Delta S = S_B$. In this case, due to $\sigma_A \gg \sigma_B$, S_{\parallel} is mainly determined by the small thermopower of the metal, and S_{\perp} is mainly given by the thermopower of the semiconductor.

The pair aluminum (A) and n-type silicon (B) was chosen for several reasons: According to the transport properties (Table I), a large anisotropy $\Delta S \sim 1,5$ mV/K approaching the thermopower of Si should result.

Table 1: Transport data for pure Al and n-type Si $(n \approx 10^{16} / \text{cm}^3)$

1	Material	$\sigma (\Omega^{-1} \mathrm{m}^{-1})$	$\lambda (\text{Wm}^{-1}\text{K}^{-1})$	S (μV/K)
	Al (A)	4·10 ⁷ [8]	232 [8]	1,66 [8]
	Si (B)	100 [9]	156 [8]	1500 [10]

Furthermore, from the constitutional diagram (Fig. 2) of Al-Si [11], it was presumed that an Al-Si multilayer structure with layers in optimum thermal and electrical contact may be pruduced by an alloying process.



Fig. 2: Phase diagram of Al-Si [11]

In Fig. 3, the response U_x , calculated from eq. 6, is plotted in dependence of thickness ratio p and tilt angle α , for an absorbed laser power $P_{abs} = 1$ W and a sample width

b = 10 mm. A maximum response is indicated for large $p \cong 20$ and $\alpha = 45^{\circ}$.

Experimental

Al-Si multilayer structures were prepared by an alloying process. First, from aluminum foils elements $10 \times 10 \text{ mm}^2$ (A) with thickness d_A were cut. From phosphorous-doped silicon (n = 10^{16} / cm³) similarly shaped elements $10 \times 10 \text{ mm}^2$ with thickness $d_B = 300 \mu \text{m}$ were prepared. The Seebeck coefficient of this Si was experimentally found to be 1,5 mV/K, in accordance to published data on arsenic doped silicon at comparable doping levels [10].

Depending on the desired thickness ratio p, d_A was varied between 15 µm (p = 20) and 300 µm (p = 1). The elements were arranged to a stack A/B/A... and heated in a tube furnace in argon atmosphere, to the eutectic temperature T_E . Reaching T_E a slight axial pressure ($\cong 1$ bar) was applied to the stack. This pressure was important to obtain large area alloying.

By trying several heating procedures it was found that, keeping the stack for 1 h at $T_{\rm E}$ a mechanically stable structure was obtained. As shown by the constitutional diagram (Fig. 2), the system Al-Si starts to form an Al-rich alloy at $T_{\rm E} = 577$ °C.

Tilted multilayer structures (Fig. 1b) were prepared by cutting a stack obliquely to the stack axis.



Fig. 3: Calculated Response U_x , as a function of tilt angle α and thickness ratio p, for $P_{abs} = 1$ W and sample width b = 10 mm.

In Fig. 4, a measurement of the thermoelectric anisotropy $\Delta S = S_{\parallel} - S_{\perp}$ as function of thickness ratio p is shown, together with a calculated $\Delta S(p)$ according to eq. 1 (line in Fig. 4). For $p \approx 20$, ΔS approaches the thermopower of Si (broken line, Fig. 4).



Fig. 4: Anisotropy versus thickness ratio. For $p \cong 20$, ΔS is approaching the Seebeck-coefficient of Si (broken line).

Tilted multilayers were obtained by cutting the stack obliquely to the layers (Fig. 1). The samples were then pasted with a heat conductive glue onto a copper mount which served as a heat sink. To maximize the light-absorption, the samples were coated with black paint. For response measurements, samples were irradiated with radiation of a cw-diode laser array (wavelength 800 nm, elliptical beam cross section) at the central sample area between the electrical contacts. Care was taken to ensure irradiation of the total sample width by aligning the long axis of the laser spot across the sample. The response in dependence of α is shown in Fig. 5 for two values of p, p = 1 and p = 20 respectively, and for a radiation power of 1 W. The response is nearly at maximum for $\alpha = 45^{\circ}$, independent of p. This is due to the fact that the thermal conductivity λ_z is nearly independent of α , due to relatively similar values of λ_A and λ_B . The lines in Fig. 5 are obtained from a calculation of the response according to eq. 2.



Fig. 5: Response of samples in dependence of tilt angle α , for p = 20 (upper curve) and p = 1 (lower curve), irradiated with a laser of power $P_{abs} = 1$ W

For p = 20 and $\alpha \approx 45^{\circ}$, a maximum sensitivity of about 0,5 mV/W results. We note that, according to eq. 2, the sensitivity depends on sample geometry. It can be increased, e. g., by concentrating the power P onto samples with smaller width. For a comparison of sensitivities (Fig. 6) samples with optimized tilt angles and comparable widths 8..10 mm were taken into account.



Fig. 6: Sensitivity of several anisotropic systems in comparison with Al-Si

Because the decay of the temperature gradient $V_z T$ occurs mainly by heat diffusion within the sample, the response time τ of the thermoelectric response upon pulsed sample heating (e.g. by pulsed laser radiation) is determined by

$$\tau_{diff} = \frac{d^2}{2D} \tag{7}$$

where d is the sample thickness and $D \sim \lambda_z$ is the thermal diffusion constant [12]. Response times τ upon pulsed TEA-CO₂-laser irradiation (pulse length 50 ns) in dependence of sample thickness are shown in Fig. 6, indicating a response time $\tau \sim d^2$. Due to large λ_z , for samples with thickness of 200 µm, response times of 30 µs were reached.



Fig. 6: Response time τ as a function of sample thickness

Conclusions

In summary, a new synthetic material with large thermoelectric anisotropy has been created. By an alloying procedure, a metal-semiconductor multilayer structure with good thermal and electrical conductivity between layers has been produced. For the first time, the very large anisotropy of metal-semiconductor multilayer structures has been demonstrated. For the system Al-Si an anisotropy

 $\Delta S \cong 1.4 \text{ mV/K}$ at room temperature has been obtained. Light sensing applications using this thermoelectric anisotropy were demonstrated.

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