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A liquid-state thermal diode

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ABSTRACT

We demonstrate a liquid-state thermal diode in which the thermal conduction changes through thermal expansion of metallic liquid to displace air. The forward (high conduction) mode occurs when the liquid spans the diode to produce a highly conducting path. The reverse mode occurs when the hot side is adjacent to air in the diode. The air provides a large thermal resistance so that the liquid metal in the reverse mode is at a lower average temperature than in the forward mode, and thus the liquid does not span the diode. With a prototype device using mercury and air in a glass tube, we demonstrate that the thermal resistance between the forward and reverse modes can differ by a factor of two, giving a rectification coefficient of about one. The rectification occurs at a variety of temperatures, and increases with the temperature difference across the diode. A figure of merit for selection of the liquid is the product of the liquid's thermal expansion coefficient and thermal conductivity. Mercury was chosen because of large values of both of these properties, and the ability to wet and dewet a surface.

1. Introduction

A thermal diode is a two terminal device that rectifies heat, that is, the heat flow, Q, is different in the forward, f, and reverse, rev, directions for the same temperature bias across the diode [1,2]. The degree of diodicity is often described by the rectification coefficient [3], r:

$$r = \frac{Q_f - Q_{rev}}{Q_{rev}} = \frac{R_{rev} - R_f}{R_f} = \frac{C_f}{C_{rev}} - 1 \tag{1}$$

where R is the thermal resistance and C is the thermal conductance. Thermal diodes have a wide variety of potential applications including thermal management in spacecraft [4], buildings [5], or microprocessor cooling [6].

The first thermal diode was a copper oxide device investigated by Starr in 1936 [7]. Subsequently, a variety of devices and mechanisms have been explored [1,3,4,6,8–15] and reviewed by Roberts and Walker [2]. Komatsu and Ito proposed a mechanism where asymmetry is introduced through asymmetry in the contact *area* of the diode to the hot and cold heat baths [14] and Hu et al. used a molecular model to describe rectification based on the diode shape [15]. Phase change thermal diodes rely on a change in R with phase, which in turn depends on the temperature [4,6,9]. They are highly effective and typically achieve $r \sim 100$. Thermal switches [16,17] are similar devices, which have a large increase in conduction at a particular temperature. In one implementation, a switch from low and high conduction occurs when the temperature is raised above the melting point of paraffin when the volume expansion during the phase transition pushes a contact into place [17,18]. Solid-state thermal rectifiers have been developed or proposed in which two solids are joined in series, where each solid has a different temperature dependence of resistivity [1,12,13]. Diodicity can also be achieved when a change in temperature is used to bring two materials into contact, thereby completing a thermal circuit. dos Santos Bernardes demonstrated a solid-state diode using thermal expansion, with $r = 0.15$ [11], and subsequently Tso et al. [10] achieved $r = 93$ using shape memory alloys and levers. One of the challenges faced when heat conducts between solids is that the solids rarely achieve molecular contact over a large area, and the remaining gaps contribute "contact resistance" in series with the solid resistance [19]. From Eq. (1), an additional contact resistance in the forward direction will reduce the rectification.

2. Working principle of the liquid-state thermal diode

Here we describe a rectifier that uses two fluids in series – one with low thermal resistivity and one with high thermal resistivity, see Fig. 1. In our prototype device, we use mercury and air as the working fluids. The advantage of liquids is that they flow easily and can repeatedly form molecular contacts with solids or other

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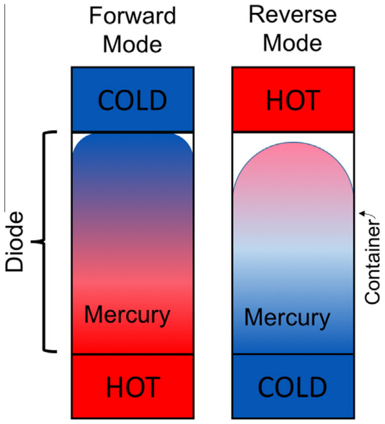

Fig. 1. Schematic showing the approximate liquid level and temperature profile in the liquid-state thermal rectifier. The hot and cold reservoirs are labeled HOT and COLD.

liquids, minimizing contact resistance. In the reverse mode, the cold side is in contact with the mercury and there is an insulating air gap between the mercury and the hot side leading to a large resistance. In the forward mode, the mercury is connected to the hot side, which causes a higher average temperature of the mercury and therefore an increase in the length of the mercury, due to thermal expansion, such that some of the air gap is eliminated. Note that the air gap does not need to be completely eliminated for rectification to occur. In the experiments described here, we operate near the limit of the largest air gap in the reverse mode such that the mercury can just contact the cold reservoir in the forward mode.

We make an approximate estimate of the rectification of such a device as follows. Assuming that the cross sectional area of the device, A, is uniform and the same in the forward and reverse modes, and the mercury meniscus is flat, the thermal resistance of the device in the reverse mode can be expressed as:

$$R_{\mathrm{rev}} = \frac{l_{\mathrm{air}}}{Ak_{\mathrm{air}}} + \frac{l_{\mathrm{liquid}}}{Ak_{\mathrm{liquid}}}, \tag{2}$$

where l and k are the length and thermal conductivity, respectively. For the case where the expansion of the liquid between the reverse and forward modes exactly matches the length of the air gap in the reverse mode, and the dimensions of the container are constant:

$$R_{\mathrm{rev}} = \frac{\alpha\Delta T' l_{\mathrm{liquid}}}{Ak_{\mathrm{air}}} + \frac{l_{\mathrm{liquid}}}{Ak_{\mathrm{liquid}}} \quad \text{and} \quad R_{\mathrm{f}} = \frac{l_{\mathrm{liquid}}}{Ak_{\mathrm{liquid}}}, \tag{3}$$

where α is the volume coefficient of thermal expansion of the liquid, and $\Delta T'$ is the difference between the average temperature of the liquid in forward and reverse modes. For calculation of the liquid resistance, we have ignored the change in length of the liquid between the forward and reverse directions because values of α are typically $<10^{-4}$/K. The rectification is then:

$$r = \frac{R_{\mathrm{rev}} - R_{\mathrm{f}}}{R_{\mathrm{f}}} \approx \frac{k_{\mathrm{liquid}}}{k_{\mathrm{air}}}\alpha\Delta T'. \tag{4}$$

Note that $\Delta T'$ is smaller than the difference in temperature between the hot and cold reservoirs, ΔT, but larger values of ΔT will cause larger values of $\Delta T'$. The figure of merit for choosing the liquid is the product, $k_{\mathrm{liquid}}\alpha_{\mathrm{liquid}}$. We fabricated our prototype from mercury in air, as the mercury–air system combines a high value of $k_{\mathrm{liquid}}\alpha_{\mathrm{liquid}}$, the potential for reversible molecular contact/separation cycling and simplicity in experimental setup and testing. Gallium ($k \sim 20$ W m^{-1} K^{-1}) in vacuum would have a superior rectification coefficient but would be more challenging to fabricate.

3. Materials and methods

A schematic of our prototype device is provided in Fig. 2. The diode was contained in a square 5 mm × 5 mm glass tube that was 20 mm long in the direction of the heat flux and had a 1.0 mm wall thickness. The volume thermal expansion coefficient of glass [20], is much less (1/20th) than mercury [21], as required for the container. The small coefficient of thermal expansion of the glass means that the diode (mercury plus container) cross section does not change, as assumed in Eq. (3). Note that the glass provides a parallel conduction path that degrades the performance of the diode. Each end of the tube was capped with a silicon wafer (0.5 mm thick) and two sensing layers: (1) aluminum shim with an imbedded thermometer (QTI Thermistors T320) and (2) a heat flux sensor (greenTEG gSKIN®-XM). The heat flux sensor had an area of 4.4 mm × 4.4 mm, which approximately matched the cross sectional area of the glass tube. A constant temperature hot or cold reservoir was at either end. Heat was provided at one end by a 50 W resistive heater with PID control (OMEGA CN32PT-220) using feedback from the thermometer. Heat was removed from the cold reservoir by recirculated water or a mixture of ethylene glycol and water. Each layer was separated by a thin layer of thermal paste (ArcticSilver 5) that facilitated heat transfer between adjacent components. The device, except the heat sink/source, was surrounded by foam insulation (Great Stuff, Dow, USA) to reduce heat losses. All measurements and handling of mercury were carried out inside a fume hood because of the toxicity of mercury vapor.

The diode consists of mercury and air. For an average change in mercury temperature of 50 °C, mercury expands by 0.3%, so for 20 mm of mercury, the air gap in the reverse direction is only about 60 μm. Prior to the thermal measurements, the height of the mercury in the diode was adjusted by mercury-filled coarse and fine syringes, and then a valve was closed to disconnect the mass of mercury in the syringes from the mass in the diode. Connection of the mercury between the two silicon wafers in the forward state

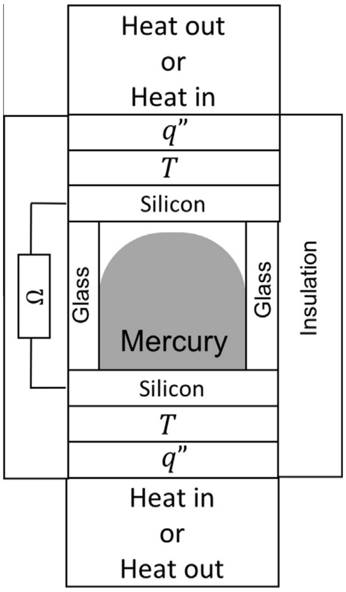

Fig. 2. Schematic of the thermal diode. The diagram is not to scale. For example, the silicon wafers are only 0.5 mm thick. Ω represents the electrical resistance measurement, T represents the temperature measurement, and q'' the heat flux measurement.

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