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Sensor Development For Single-Photon Thermoelectric Detectors

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Abstract. As we reported earlier [1], thermoelectric detectors can be competitive as non-
dispersive energy resolving focal-plane instruments in X-ray/UV spectrum. The first generations
of prototype devices demonstrated the viability of detector design and provided good agreement
between theoretical expectations and experimental data. These devices exploited sensors made
of gold with a small fraction of iron impurity. To get the projected high resolution one needs
another type of material, namely, lanthanum-cerium hexaborides. We report on the first
experimental tests of the feasibility of lanthanum-cerium films as sensor materials. Progress with
thin films of these materials argues for the success of these thermoelectric detectors.

1. INTRODUCTION

When a photon is absorbed in material its energy is converted to excitations in the
material. The photon's energy quickly appears as temperature excess near the
absorption site. Measuring this excess is one way of determining what energy the
photon had. Because of an effect that has been well known since the early nineteenth
century, — thermoelectricity — the temperature difference between the photon
absorption site and its surroundings will have associated with a measurable (transient)
voltage difference and it is possible that this difference can be exploited for photon
characterization. A priori, there is no way to be certain that it is not better than other
methods; one must investigate it.

2. EXPERIMENTAL RESULTS WITH GOLD-IRON SENSORS

Figure 1 shows the crucial portion of the device associated with the two absorbing
pixels and the thermoelectric sensor connecting them. It also indicates the material
and geometrical parameters. In operation the device is cooled to temperatures below
1K and laser pulses are used to determine its response. The usual scheme for
measurements is shown in Fig. 2 (a). To assure that only one pixel is illuminated in the pulse a reflecting foil, as shown by Fig. 2(b) screens one half of the detector. Thin and narrow superconducting leads (detectable in Fig. 1) connect the two-pixel region of the device to the signal acquisition circuitry, schematically described in Fig. 3(a). The typical device output is shown in Fig. 3(b).

Devices built using gold with controlled iron impurities were the natural choice for the model validation phase. Fundamental characteristics of the output such as the amplitude, signal duration, signal/noise ratio agree well between the theory and the experiment. The energy resolution obtained with gold/iron devices was about 500 eV at 6 keV energy deposited. In principle, it was possible to pursue more than an order
of magnitude higher resolution with this material, by adjusting the iron concentration in gold, as well as the geometrical parameters of the sensor and material parameters of the absorber. However this is not a development path that would lead to the highest performance devices. The model predicts spectral resolution to be [1]:

\[ \Delta E_{\text{FWHM}} = 2.35 \left( 2 k_B T^2 C_{\text{abs}} \left( 1 + L_0 / S^2 \right) \right)^{1/2}. \]  

where \( S \) is Seebeck’s coefficient \( S \) for the sensor material. Other quantities in Eq. (1) are: \( C_{\text{abs}} \) – the absorber heat capacity, \( T \) – the operational temperature, \( L_0 \) – the Lorenz number. The detector is not optimized unless the values of \( S \) are higher than (or at least close to) 100 \( \mu \text{V/K} \) at operational temperatures. In the best case gold/iron material can have \( S \) about 10 \( \mu \text{V/K} \), which is not sufficient. Extensive search in the literature revealed another Kondo-metal, Lanthanum Hexaboride with partial (1%) substitution of Lanthanum by Cerium (Fig. 4(a)). We investigated whether the properties of this material agreed with assumptions made in our theoretical modeling, in particular, how closely the Wiedemann-Franz relation \( (k_e/\sigma; \text{see Ref. 1}) \) is followed (Fig. 4(b)). These figures argue strongly that the requirements have a chance to be met.

3. KONDO-MECHANISM IN HEXABORIDE FILMS

Since the basic constituent of this development is thermoelectric sensors, we have focused our recent experimental efforts on the films of the candidate material \( \text{La}_{0.99}\text{Ce}_{0.01}\text{B}_6 \) with best known properties for the detector application. As in Au(Fe) case, in this material, the high value of Seebeck coefficient is due to the Kondo-mechanism. For \( \text{La(Ce)B}_6 \) the resistance of the bulk samples behaves as shown in Fig. 5, left. This is the simplest measurement one can perform to confirm the presence of the Kondo mechanism and check the quality of the film. In films, there is typically higher residual contribution (temperature independent background). The

FIGURE 4. (a) Seebeck coefficients of \( \text{La}_{0.99}\text{Ce}_{0.01}\text{B}_6 \) (Ref. 2) and optimized Au(Fe) (Ref. 3). The hexaboride is obviously preferable. (b) Below 3K the Wiedemann-Franz law holds not only in \( \text{LaB}_6 \), but also in \( \text{CeB}_6 \) [4]. No Lorenz number data exist yet for \( \text{La}_{0.99}\text{Ce}_{0.01}\text{B}_6 \).
FIGURE 5. Kondo-resistivity in bulk La(Ce)B6 samples should have a minimum at about T=20K (Ref.5). This figure documents our experimental data for 1μm thick La0.99Ce0.01B6 films (the constant background subtracted, see text).

greater the purity and crystallinity of the sample, the lower the background. We used e-beam deposition system to deposit La0.99Ce0.01B6 onto heated (up to 950°C) substrates from stoicheometric targets (99.9% purity). Multiple technological improvements very recently yielded films that are only a factor of two higher in room-temperature resistance than bulk samples described in the literature. We are now at the stage of measurement of these samples at low temperatures. Figures 5 and 6 are the first demonstration anywhere that the Kondo effect is seen in thin films of hexaborides. This is an interesting result in materials science for thermoelectrics, but it is crucial and enormously encouraging for our detector development. Seebeck coefficient in thin films will be measured in the next step.

FIGURE 6. Residual resistivity and Kondo behavior of recently deposited and tested films. The value for the bulk material is shown for comparison. Thin films are rapidly approaching the bulk limit as our technique improves.

REFERENCES