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**Nanoscale, phonon-coupled calorimetry with sub-attojoule/kelvin resolution**

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We have developed a nanofabricated calorimeter for ultrasensitive heat capacity measurements upon externally-affixed (phonon-coupled) samples at low temperatures. For a 5s measurement at 2K, we demonstrate unprecedented resolution of  $C$  of 0.5 aJ/K (approximately 36,000 kB). This sensitivity enables, for example, possible applications such as heat capacity measurements upon zeptomole-scale samples and upon adsorbates at extremely minute coverage. We describe the configuration and operation of these devices, and demonstrate their operation by measuring an adsorbed He4 film with maximum resolution of approximately  $3 \times 10^{-5}$  monolayers upon the minute device surface area,  $A$  approximately  $1.2 \times 10^{-9}$  m<sup>2</sup>.