

Thermodynamic evidence for valley-dependent density of states in bulk bismuth

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Over the last seven years, bismuth has attracted new attention because of its unusual properties in the presence of a strong magnetic field. **(i)** Nernst effect measurements, well above the quantum limit, found features unexpected in the single-particle picture [1]. Even more striking, **(ii)** Torque magnetometry studies up to 31 T [2] showed a structure in magnetization with hysteresis suggesting a first order correlation-induced phase transition. Most recently, **(iii)** measurements of the magnetoconductivity tensor detected a spontaneous loss of the threefold rotational symmetry of the underlying lattice, as reported by Zhu *et al.* [3]. **(iv)** However, the relevance of internal strain could not be ruled out [4].

To this lively debate, we bring our high-resolution angle-dependent magnetostriction measurements of bismuth. The magnetostriction coefficient provides a direct thermodynamic measurement of changes in the carrier density: peak positions in the (field,angle) plane can be mapped to the Landau level spectra of electron and hole valleys, and compared with theory [5]. Contributions from electrons and holes are resolved with exceptional clarity.

Our main result is that we find compelling evidence for spontaneous valley imbalance. In contrast to transport **(iii)**, the thermodynamic probe employed in this study directly couples to the density of states. Our experiments establish for the first time that while two distinct valleys keep identical Landau spectra, they present a different density of states at the Fermi level [6]. These rules out the possibility that uncontrolled strain is responsible for lifting the valley degeneracy, and sets boundaries on the possible theoretical scenarios proposed in **(iv)**.

Another important result of our thermodynamic study is that it reveals no trace of hysteresis and does not detect any unexpected additional features above the quantum limit. Therefore we can clearly exclude the existence of correlation induced thermodynamic phases, which were proposed in **(i)** and **(ii)** to explain the ultraquantum phenomena.

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[5] J. Alicia and L. Balents, *Phys.Rev. B* **79**, 241101 (2009).

[6] R.Küchler *et al.*, *Nature Materials* 3909, 10.1038 (2014).