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

LUCIA STEINKE, Brookhaven National Laboratory, 11973 Upton, NY

A moderate magnetic field confines both hole-like and electron-like carriers of semi-metallic bismuth to their lowest Landau levels. In contrast to holes, which are ordinary quasi-particles, electrons in bismuth are described by the Dirac Hamiltonian with a band mass becoming a thousandth of the bare electron mass along one crystalline axis. These Dirac electrons can occupy each of the three rotationally equivalent elongated ellipsoids of the Fermi surface. The valley degeneracy offers electrons an additional degree of freedom, a subject of recent attention. Here, we employ magnetostriction to map the angle-resolved Landau spectrum and quantify the number of electrons in each valley for a magnetic field slightly tilted off the trigonal axis. Unlike transport measurements, magnetostriction provides a thermodynamic probe that directly couples to the density of states: resonances in the magnetostriction coefficient can be linked to the evacuation of Landau levels with increasing field, and electron and hole spectra are distinguished by tracing the angle dependence of the magnetostriction peaks. We find that while the electron valleys remain identical in their spectrum, they substantially differ in their density of states at the Fermi level, with an $\approx 20\%$ difference in the peak height for two different electron valleys. This experimental observation establishes that, even in the absence of internal strain, the electron fluid does not keep the rotational symmetry of the lattice, at low temperature and high magnetic field. The valley imbalance is found to be restricted to electrons in the immediate vicinity of the Fermi level. This effect, reminiscent of Coulomb pseudo-gap in localized electronic state, emerges as the most striking departure from the non-interacting picture of electrons in bulk bismuth in the vicinity of the quantum limit.