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Nanothermodynamics in the strong coupling regime[1]

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In macroscopic thermodynamics, energy gained by a system is lost by its surroundings (or vice-versa), in accordance with the first law of thermodynamics. However, if the system-environment interaction energy cannot be neglected – as in the case of a microscopic system such as a single molecule in solution – then it is not immediately clear where to draw the line between the energy of the system and that of the environment. To which subsystem does the interaction energy belong? I will describe a microscopic formulation of both the first and second laws of thermodynamics that applies to this situation. In this framework, seven key thermodynamic quantities – internal energy, entropy, volume, enthalpy, Gibbs free energy, heat and work – are given precise microscopic definitions, and the first and second laws are preserved without requiring corrections due to finite system-environment coupling. Furthermore, these definitions reduce to the usual ones in the limit of macroscopic systems of interest. This condition establishes that a unifying framework can be developed, encompassing stochastic thermodynamics at one end and macroscopic thermodynamics at the other. A central element of this framework is a thermodynamic definition of the volume of the system of interest, which converges to the usual geometric definition when the system is large.

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