

THE THERMODYNAMIC PROPERTIES OF $^{138,139}\text{La}$ NUCLEAR SYSTEMS

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The investigation of thermodynamic properties of nuclei as a function of excitation energy is one of the challenges in nuclear physics. In this work, we have, for the first time, extracted the entropy, entropy excess, nuclear temperature, and heat capacity of the $^{138,139}\text{La}$ nuclei as a function of excitation energy from their experimental nuclear-level densities. We observed that the odd-odd ^{138}La has a higher entropy due to the additional unpaired neutron. In particular, it has an average entropy excess of $2.3 \pm 0.5 k_{\text{B}}$, which corresponds to the 10 times higher nuclear-level density due to the last unpaired neutron in ^{138}La . The nuclear temperature and heat capacity show evidence for the sequential breaking of Cooper nucleon pairs and as a result, we observed approximately constant temperatures of 0.7 ± 0.3 and 0.8 ± 0.2 MeV in the quasi-continuum of ^{138}La and ^{139}La , respectively.

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1. Introduction

In nuclear reactions, nuclei can be excited to various excitation energy, E_x , regions such as discrete energy levels and quasi-continuum. As the excitation energy increases to the quasi-continuum region, the level spacing decreases and the width of the quantum states becomes wider. At such high excitation energies, it is very difficult to resolve individual nuclear-energy levels using the present experimental energy resolution. Hence, the electromagnetic properties of nuclei that are excited to the quasi-continuum energy region are best described using average quantities such as the nuclear-level density $\rho(E_x)$. There are various analytical and microscopic theoretical models of $\rho(E_x)$. One of the popular analytical expressions of the nuclear-level density is the back-shifted Fermi gas model [1], which is the parametrization

of the original Fermi gas model of Bethe [2]. This allows the calculation of ρ as a function of E_x and spin J . The ρ can also be measured experimentally, as a function of E_x , using advanced techniques such as the Oslo Method [3], which is based on the primary γ spectra of a nucleus of interest.

The nuclear-level density can be used to compute numerous observables. For instance, it can be used as the critical input data to calculate neutron capture cross sections within the Hauser–Feshbach model [4]. It can also be used as a partition function to investigate the thermodynamic properties of nuclei, based on the microcanonical ensemble model. This is the main focus area of this paper. This area of research has attracted many theoretical and experimental studies [5–9]. The results of these studies have revealed fine structures of the nuclear temperature and heat capacity, among other observations. For instance, the experimental results yielded negative heat capacity and the decreasing nuclear temperature at excitation energies that correspond to the quenching of Cooper nucleon–nucleon pairs. Some nuclei such as ^{198}Au show a constant temperature at $E_x > 2\Delta$, where Δ is the pairing gap parameter. On the other hand, the temperature of nuclei such as ^{239}U and ^{196}Pt shows small oscillations about a constant temperature at excitation energies beyond the location of the bump-like structure that occurs at $E_x \approx 2\Delta$ [5, 6]. These oscillations have not been fully understood, although they are probably due to the sequential melting of the multiple Cooper pairs. Furthermore, the theoretical investigations have predicted a peak-like structure in the proton entropy excess, at $E_x < 1$ MeV, that is due to the pairing re-entrance phenomenon, but this has not been observed experimentally [8].

In this work, we investigated, for the first time, the thermodynamic properties of the $^{138,139}\text{La}$ nuclei, to obtain more insight into these many-body systems. In particular, we extracted the entropy, entropy excess, nuclear temperature, and heat capacity of the $^{138,139}\text{La}$ nuclei, from their experimental nuclear-level density data.

2. Methods

The experimental nuclear-level density, $\rho(E_x)$, data of Ref. [10] were used in this work as the input data for extraction of the thermodynamic properties of the $^{138,139}\text{La}$ nuclei. These are depicted in figure 1. In particular, at each excitation energy, a nucleus is an isolated system with the fixed total energy. Its volume and number of nucleons also remain constant in the excitation energy below the neutron threshold. Hence, a nuclear system is a microcanonical ensemble and its entropy at each excitation energy is given by

$$S(E_x) = k_B \ln(W(E_x)), \quad (1)$$

where $W(E_x)$ is the number of accessible states, k_B is the Boltzman constant, and $W(E_x) \propto (2J + 1)\rho(E_x)$. The $(2J + 1)$ factor is the spin degeneracy of a specific quantum state with excitation energy E_x and spin quantum number J .

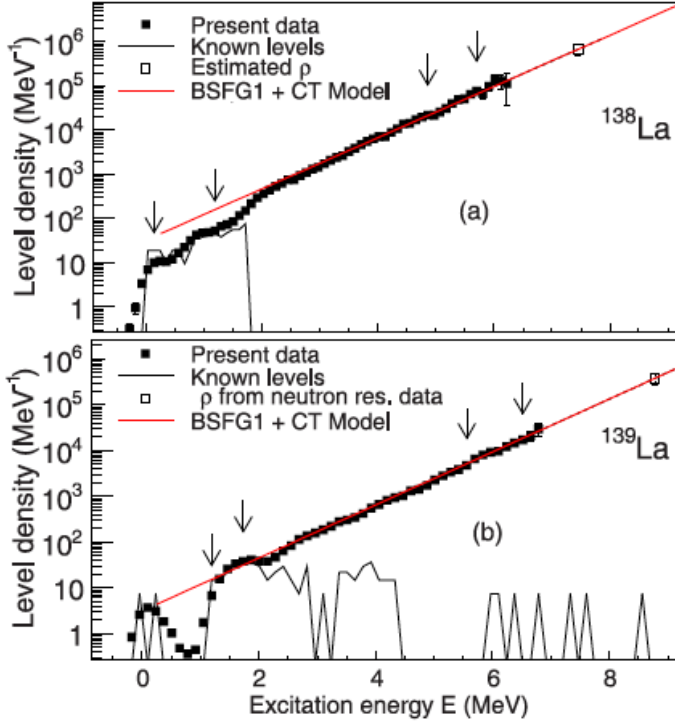


Fig. 1. Experimental nuclear-level density of ^{138}La (upper panel) and ^{139}La (lower panel) [10]. Black solid squares are experimental data measured at the Oslo cyclotron laboratory. Black solid lines are $\rho(E_x)$ obtained using the known discrete levels from Ref. [11]. Open black squares are $\rho(E_x)$ calculated at the neutron separation energies of $^{138,139}\text{La}$. Red lines are $\rho(E_x)$ computed with the back-shifted Fermi gas model coupled with constant temperature model.

However, the experimental method used to measure the nuclear-level density shown in figure 1 cannot provide any data about the spin degeneracy of magnetic substates. Thus, we can only extract the reduced entropy, which is given by

$$S(E_x) = \ln \left(\frac{\rho(E_x)}{\rho_0} \right), \quad (2)$$

in units of k_B , where ρ_0 is the normalization constant, which ensures that the third law of thermodynamics is satisfied.

It also follows that the nuclear temperature and heat capacity as a function of excitation energy are, respectively, given by

$$T(E_x) = \left(\frac{\partial S(E_x)}{\partial E_x} \right)^{-1} \quad (3)$$

and

$$C_v = \left(\frac{\partial T(E_x)}{\partial E_x} \right)^{-1}. \quad (4)$$

3. Results and discussion

This section provides the discussion of the thermodynamic quantities of $^{138,139}\text{La}$ that were obtained using the input experimental nuclear-level density data shown in figure 1 and equations (2), (3), and (4).

Figure 2 shows the entropy, $S(E_x)$, of both isotopes as a function of excitation energy. The $S(E_x)$ of the odd-odd ^{138}La is higher than that of the odd-even ^{139}La . In particular, it ranges from $0.87 k_B$ to $10 k_B$ for the former, while it ranges from $0.050 k_B$ to $8.8 k_B$ for the latter. We also observe that the $S(E_x)$ of ^{139}La approaches $-\infty$ in the 0.20 MeV to 1.2 MeV energy region. This is because this nucleus has no energy levels between 165.9 keV and 1209.0 keV states [11]. Hence, in this E_x region, there is no entropy.

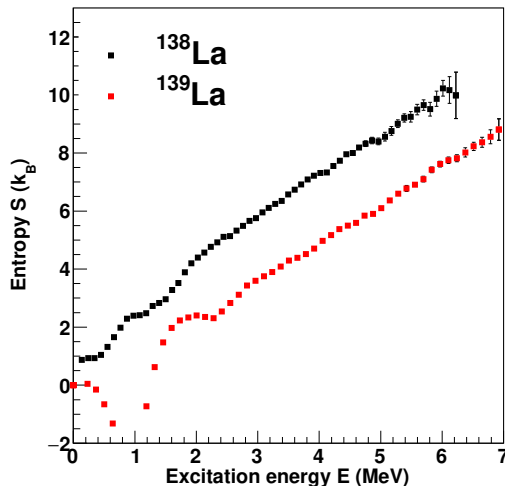


Fig. 2. The entropy of ^{138}La and ^{139}La .

Figure 3 shows the entropy excess between ^{138}La and ^{139}La due to the unpaired neutron in the ^{138}La nucleus. It is observed that, on average, the entropy excess is $\Delta S = 2.3 \pm 0.5 k_B$ for $E_x > 2.4 \text{ MeV}$. This value was

obtained by computing the average of all entropy excess values in the excitation energy region above 2.4 MeV. Its uncertainty was estimated through error-propagation using the uncertainties of all data points in the $E_x > 2.4$ MeV region. It corresponds to the $n = \exp(\Delta S) = \exp(2.3) \approx 10$ times higher nuclear-level density due to the last unpaired neutron in ^{138}La .

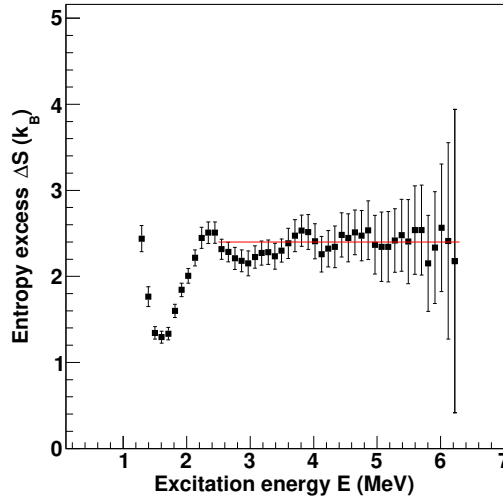


Fig. 3. The entropy excess of ^{138}La . The red horizontal line is the average entropy excess.

Furthermore, the nuclear temperature was extracted using equation (3) for both isotopes and is depicted in figure 4. The temperature of ^{138}La cannot be extracted at $E_x < 0.6$ MeV because the entropy is flat in this energy region due to the ground band, which means that $\partial S(E_x)/\partial E_x$ in equation (3) is undefined. An observation similar to this one was also reported on the $^{237,238,239}\text{U}$ nuclei [6]. Similarly, the temperature of ^{139}La nucleus was also not determined at excitation energies below 1.2 MeV due to the lack of energy levels. However, above the energies of 0.6 MeV and 1.2 MeV, the temperatures of these nuclei increase up to 0.75 MeV and 1.2 MeV in the $E_x = 0.8\text{--}1.2$ MeV and $E_x = 1.3\text{--}2.1$ MeV, respectively. Above these E_x regions, the nuclear temperature decreases and reaches minima at $E_x = 1.8$ MeV and $E_x = 2.8$ MeV for ^{138}La and ^{139}La , respectively. This decrease in nuclear temperature with the increase in energy of the systems is due to the melting of the first Cooper nucleon pairs. This is also evident in the caloric curve, in figure 5, which shows that heat capacity is negative at $E_x = 1.2\text{--}1.8$ MeV and $2.1\text{--}2.8$ MeV for the ^{138}La and ^{139}La , respectively. These results are consistent with other studies [6]. The observation of systems that cool down although their energy increases has been

reported in many other objects such as stars and atomic clusters [12–16]. This phenomenon is well understood using the Bardeen–Cooper–Schrieffer (BCS) theory [17].

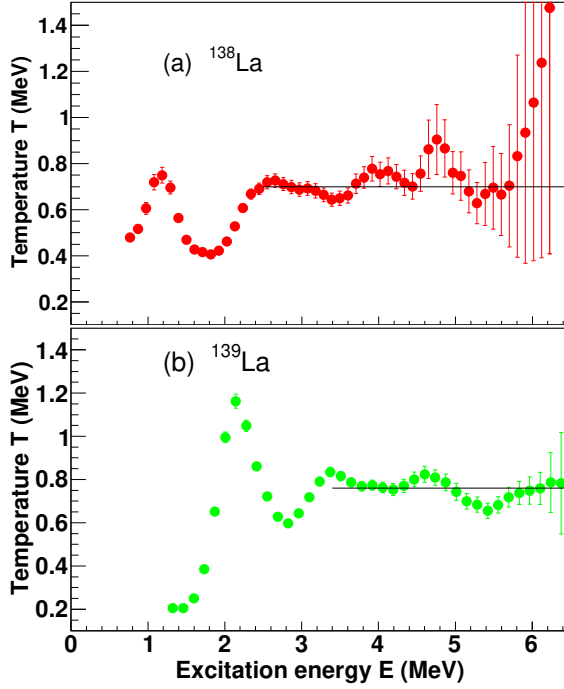


Fig. 4. The nuclear temperature of (a) ^{138}La and (b) ^{139}La . The black horizontal lines are the average temperatures.

Figure 4 also shows that the nuclear temperatures of ^{138}La and ^{139}La , respectively, increase in the 1.8–2.5 MeV and 2.8–3.4 MeV excitation energy regions. This is due to the latent pairing energy being released to the isolated nuclear systems. Above these energies, the temperature remains relatively constant around the average temperatures 0.7 ± 0.3 MeV and 0.8 ± 0.2 MeV for ^{138}La and ^{139}La , respectively, although there are small-amplitude oscillations about these temperatures. These average temperatures were obtained using the same method that was used to calculate the average entropy excess, which is discussed above. The small-amplitude oscillations and the corresponding negative heat capacities (see figure 5) suggest that there is a consecutive breaking of multiple Cooper nucleon pairs. Furthermore, the constant nuclear temperature, in the quasi-continuum region, has been reported in many other nuclear systems in the literature [5, 18].

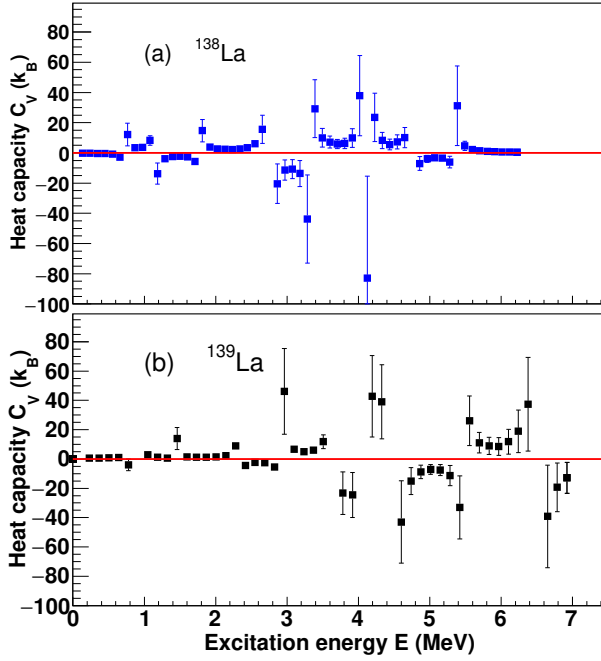


Fig. 5. The heat capacity of (a) ^{138}La and (b) ^{139}La . The horizontal red line represents the heat capacity of zero.

4. Summary and conclusions

The experimental nuclear-level density data of the $^{138,139}\text{La}$ nuclei were used to extract their entropy, nuclear temperature, heat capacity, and the entropy excess of the odd-odd ^{138}La as the function of excitation energy. The results showed that the odd-odd ^{138}La has a higher entropy due to the extra unpaired neutron. In particular, the average entropy excess between these isotopes is $\Delta S = 2.3 \pm 0.5 k_B$, and it corresponds to the 10 times higher nuclear-level density of ^{138}La due to its unpaired neutron. Furthermore, the nuclear temperature and heat capacity show a rise and fall in the 0.8–1.8 MeV and 1.3–2.8 MeV excitation energy regions. These are due to the breaking of the first Cooper nucleon pairs. It is also observed that the $^{138,139}\text{La}$ isotopes have approximately constant temperatures, of which the average values are, respectively, 0.7 ± 0.3 MeV and 0.8 ± 0.2 MeV in the quasi-continuum. This constant nuclear temperature results from the sequential melting of multiple Cooper pairs, as seen in the caloric curve and nuclear temperature.

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REFERENCES

- [1] A. Gilbert, A.G.W. Cameron, «A composite nuclear-level density formula with shell corrections», *Can. J. Phys.* **43**, 1446 (1965).
- [2] H.A. Bethe, «An Attempt to Calculate the Number of Energy Levels of a Heavy Nucleus», *Phys. Rev.* **50**, 332 (1936).
- [3] A. Schiller *et al.*, «Extraction of level density and γ strength function from primary γ spectra», *Nucl. Instrum. Methods Phys. Res. A* **447**, 498 (2000).
- [4] W. Hauser, H. Feshbach, «The Inelastic Scattering of Neutrons», *Phys. Rev.* **87**, 366 (1952).
- [5] F. Giacoppo *et al.*, «Level densities and thermodynamical properties of Pt and Au isotopes», *Phys. Rev. C* **90**, 054330 (2014).
- [6] M. Guttormsen *et al.*, «Constant-temperature level densities in the quasicontinuum of Th and U isotopes», *Phys. Rev. C* **88**, 024307 (2013).
- [7] B. Dey *et al.*, «S-shaped heat capacity in an odd–odd deformed nucleus», *Phys. Lett. B* **789**, 634 (2019).
- [8] B. Dey *et al.*, «Proton entropy excess and possible signature of pairing reentrance in hot nuclei», *Phys. Lett. B* **819**, 136445 (2021).
- [9] P. Roy *et al.*, «Nuclear level density and thermal properties of ^{115}Sn from neutron evaporation», *Eur. Phys. J. A* **57**, 48 (2021).
- [10] B.V. Kheswa *et al.*, « $^{137,138,139}\text{La}(n,\gamma)$ cross sections constrained with statistical decay properties of $^{138,139,140}\text{La}$ nuclei», *Phys. Rev. C* **95**, 045805 (2017).
- [11] <https://www.nndc.bnl.gov/>
- [12] W. Thirring, «Systems with negative specific heat», *Z. Phys.* **235**, 339 (1970).
- [13] D. Lynden-Bell, «Negative specific heat in astronomy, physics and chemistry», *Physica A (Amsterdam)* **263**, 293 (1999).
- [14] M. Bixon, J. Jortner, «Energetic and thermodynamic size effects in molecular clusters», *J. Chem. Phys.* **91**, 1631 (1989).
- [15] P. Labastie, R.L. Whetten, «Statistical thermodynamics of the cluster solid–liquid transition», *Phys. Rev. Lett.* **65**, 1567 (1990).
- [16] M. Schmidt *et al.*, «Negative Heat Capacity for a Cluster of 147 Sodium Atoms», *Phys. Rev. Lett.* **86**, 1191 (2001).
- [17] J. Bardeen *et al.*, «Theory of Superconductivity», *Phys. Rev.* **108**, 1175 (1957).
- [18] E. Melby *et al.*, «Thermal and electromagnetic properties of ^{166}Er and ^{167}Er », *Phys. Rev. C* **63**, 044309 (2001).