

# Gold cluster radiation from thermally populated excited electronic states

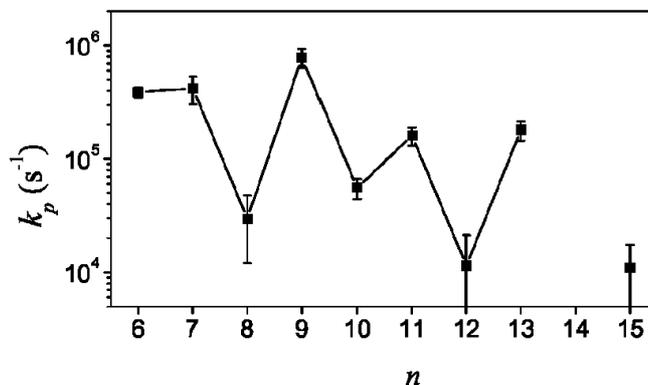
K. Hansen<sup>\*1</sup>, P. Ferrari<sup>2</sup>, E. Janssens<sup>2</sup>, P. Lievens<sup>2</sup>

1. Center for Joint Quantum Studies and Department of Physics, Tianjin University, 92 Weijin Road, Tianjin 300072, China

2. Laboratory of Solid State Physics and Magnetism, KU Leuven, 3001 Leuven, Belgium

Small positively charged gold clusters have been found to emit thermal radiation at a very high rate, with time constants ranging from one to 35  $\mu\text{s}$  for  $\text{Au}_n^+$  ( $n = 6 - 13, 15$ ) [1]. Photon emission time constants of this magnitude are only consistent with emission of thermally excited electronic states. Direct detection of the emitted photons in the analogous process in small carbon clusters have confirmed the nature of this decay [2]. For sizes  $n = 14, 16 - 20$  the radiation occurs on much longer time scales, beyond the reach of the reflectron Time-of-Flight mass spectrometer used in this study.

The clusters were produced neutral in a laser ablation source [3] and excited, ionized and strongly fragmented by laser pulses of either 20 mJ at 355 nm or 140 mJ at 532 nm wavelengths, with no change in the observed quantities. The extensive fragmentation excludes long-lived states as the origin of the photons and allows an analysis based on broad internal clusters energy distributions. Radiative cooling was inferred from the suppression of the metastable evaporation between mass selection and analysis in the reflectron. Fig. shows the results of the measurements.



**Fig. 1:** The fitted values of the photon emission rate constant,  $k_p$ . The uncertainties are 1- $\sigma$  values. The values for  $n = 14, 16 - 20$  are consistent with zero.

The measured data combined with the Thomas-Reiche-Kuhn dipole transition sum rule limit the possible excited state energies, and gives a minimum oscillator strength required. The upper limits of the state energies are given all around 1.5 eV. The necessary oscillator strengths are in all cases below  $f = 1$  and usually less than  $f = 0.3$ , corresponding to 0.3 electron out of the  $n - 1$  valence electrons.

These and similar results for niobium[4], silicon[5], and carbon[6],[7],[8] suggest that this dissipation mechanism from highly excited clusters may be more widespread than previously anticipated.

## References

- [1] K. Hansen, P. Ferrari, E. Janssens, and P. Lievens, *Phys. Rev. A* **96** 022511 (2017)
- [2] Y. Ebara, T. Furukawa, J. Matsumoto, H. Tanuma, T. Azuma, H. Shiromaru, and K. Hansen, *Phys. Rev. Lett.* **117** 133004 (2016)
- [3] W. Bouwen, P. Thoen, F. Vanhoutte, S. Bouckaert, F. Despa, H. Weidele, R.E. Silverans, and P. Lievens, *Rev. Sci. Instrum.* **71** 54-58 (2000)
- [4] K. Hansen, Y. Li, V. Kaydashev, and E. Janssens, *J. Chem. Phys.*, **141** 024302 (2014)
- [5] P. Ferrari, Ewald Janssens, Peter Lievens, and Klavs Hansen, *J. Chem. Phys.* **143** 224313 (2015)
- [6] K. Hansen and E.E.B. Campbell, *J. Chem. Phys.* **104** 5012 (1996)
- [7] J. U. Andersen, C. Brink, P. Hvelplund, M. O. Larsson, B. B. Nielsen, and H. Shen, *Phys. Rev. Lett.* **77** 3991 (1996)
- [8] G. Ito, T. Furukawa, H. Tanuma, J. Matsumoto, H. Shiromaru, T. Majima, M. Goto, T. Azuma, and K. Hansen, *Phys. Rev. Lett.* **112** 183001 (2014)

\*Corresponding author: klavshansen@tju.edu.cn