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Classification of Metal Nanoclusters Using Convolutional Neural Networks

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Catalysis happens only at the surface of materials, this makes nanoparticles of particular interest in the field of catalysis because of their high surface-to-volume ratio. The exact atomic structure of nanoparticle surfaces is of particular importance in catalysis, and the expression of surface facets is largely governed by their overall structure. Typically, small metal nanoparticles will take one of three major structural isomers: decahedron, icosahedron or cuboctahedron (Figure 1). Determination of the structural isomer of a nanoparticle can be performed using high-angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) [1,2].

In this study, we are investigating size-selected gas-condensation magnetron sputtered clusters [3]. In particular, we are interested in so-called "magic number" nanoparticles, which have a complete closed outer "shell" of atoms [1]. Previous studies have attempted to manually count the number of each structural isomer, to calculate the relative abundance of each structure and therefore determine their relative potential energies [1,2]. This is of interest to understand the magnetron conditions required to make specific surface facets for catalytic applications.

Manual identification of such nanoparticles is a time-consuming process, and therefore we turn to the booming field of scientific automation in machine learning. Machine learning has become popular in electron microscopy for solving a variety of problems [4]. Here, we turn these techniques to the classification problem of nanocluster identification. Specifically, we use a convolutional neural network (CNN), a class of machine learning algorithm that can be trained to recognize image features [5].

In our work, a convolutional neural network has been trained using HAADF-STEM images of particles, simulated using the plane-wave reciprocal-space interpolated scattering matrix (PRISM) algorithm (Figure 1 d-u), to recognize the different shapes and patterns of nanocluster images acquired in HAADF-STEM [6]. The neural network can be used to rapidly determine the proportion of different particle isomers. The speed improvements afforded by a neural network approach will allow us to process videos of atomic clusters, determining their structure in each frame, as demonstrated using a manual classification approach in Figure 2. Through video-rate imaging at a range of temperatures we will be able to determine branching ratios and therefore calculate the energy barriers between the different structural isomers. This will provide additional information on the applicability of different clusters for use at high temperatures, in terms of which surface facets will persist under reaction conditions.

The trained neural network will be made available for anyone to use via its incorporation in to the open-source software package ParticleSpy [7]. This will allow anyone with STEM images of nanoparticles to use the same technique to analyse the shape of small nanoparticles. This work also has extensions in the identification of particle orientations, in-depth 3D structure determination/mapping, and image segmentation.

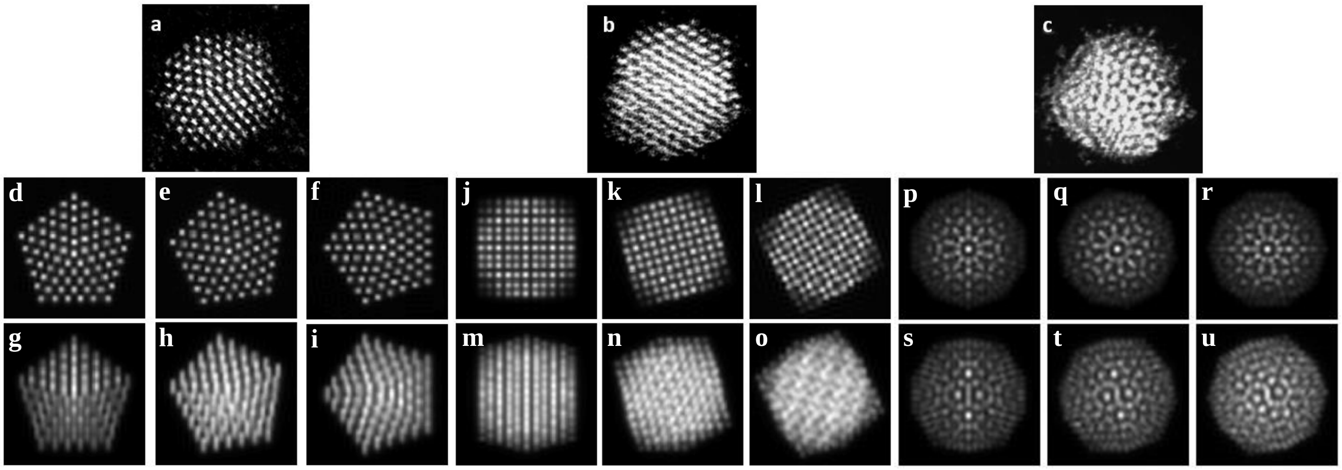

Figure 1. (a) HAADF-STEM image of decahedral nanocluster. (b) HAADF-STEM image of cuboctahedral (fcc) nanocluster. (c) HAADF-STEM image of icosahedral nanocluster. (d-i) Simulations of decahedra used for identification of decahedral isomer motifs. (j-o) Simulations of cuboctahedra used for identification of cuboctahedral isomer motifs. (p-u) Simulations of icosahedra used for identification of icosahedral isomer motifs.

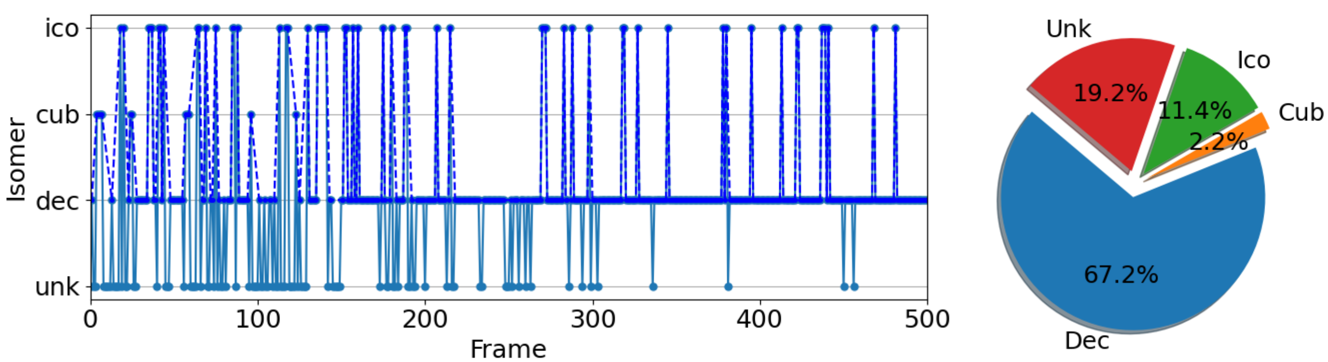

Figure 2. Manual classification results from a HAADF-STEM image series of a single 309 atom Au cluster at room temperature. Labels correspond to icosahedral (ico), decahedral (dec), cuboctahedral (cub), and unknown/amorphous (unk).

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