

Interpretability of Low-Dose HRTEM Images of Supported Metal Nanoparticles

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Modern transmission electron microscopy (TEM) produces large amounts of data, particularly when recording video datasets under active conditions (*in situ*). Manual processing of such datasets is a time-consuming and introduces potential for errors due to operator bias. In addition to time costs, analysis of these datasets is also difficult as they are typically recorded at low electron dose rates in order to minimize the effect of sample-beam interactions. Under these conditions, datasets often suffer from low signal-to-noise ratio (SNR). Recently, machine-learning has been suggested as a viable solution to analyze such datasets [1]. In this paper, we will evaluate the interpretability of low SNR datasets prior to machine-learning based analysis.

An increased SNR is often achieved by increasing the electron dose rate or by increasing the acquisition time – the first is preferred over the latter to capture dynamic events in the nanoparticle. Figure 1 shows a series of HRTEM images acquired at increasing dose rates but with constant exposure time, resulting in improved SNR. A consequence of increased dose rate can be irreversible changes of the sample or an increase of the atom mobility (Fig. 1(d)), hence low electron dose rates are desirable. However, this comes at the cost of decreased SNR and thus potentially challenging the interpretability of the images.

So how much can the electron dose rate be decreased while still allowing us to produce analyzable data while avoiding irreversible sample damage and maintaining high temporal resolution? Traditionally, the definition of SNR is $\mu(I)/\sigma(I)$, where $\mu(I)$ and $\sigma(I)$ are the mean and standard deviation of the signal I , respectively [2]. The Rose criterion states that at an $\text{SNR} \geq 5$ an image is interpretable enough to observe features with sufficient accuracy [3,4]. As this might primarily apply for images dominated by amplitude contrast, it may not be suitable for images containing predominantly phase contrast such as high-resolution TEM (HRTEM) images.

In this study, we evaluate different approaches to estimate the analyzability and interpretability of HRTEM images. Using a model system consisting of gold nanoparticles supported on cerium dioxide [5,6], we established several relevant image quality measurements and demonstrated how a machine learning based data analysis performs as a function of these parameters.

An initial measurement of SNR is shown in Fig. 2. where areas of measurements are indicated on Fig. 2(a). The traditional SNR is shown in Fig. 2(b), compared to an SNR model including the variance of the signal in the nanoparticle compared to the variance of the noise in the surrounding high vacuum area (Fig. 2(c)) and a revised SNR model [4] including the mean squared difference between the nanoparticle signal and the background noise. The newer models also indicate strong fluctuations over time in the SNR as shown in Fig. 2(e)-(f), especially at very large dose rates.

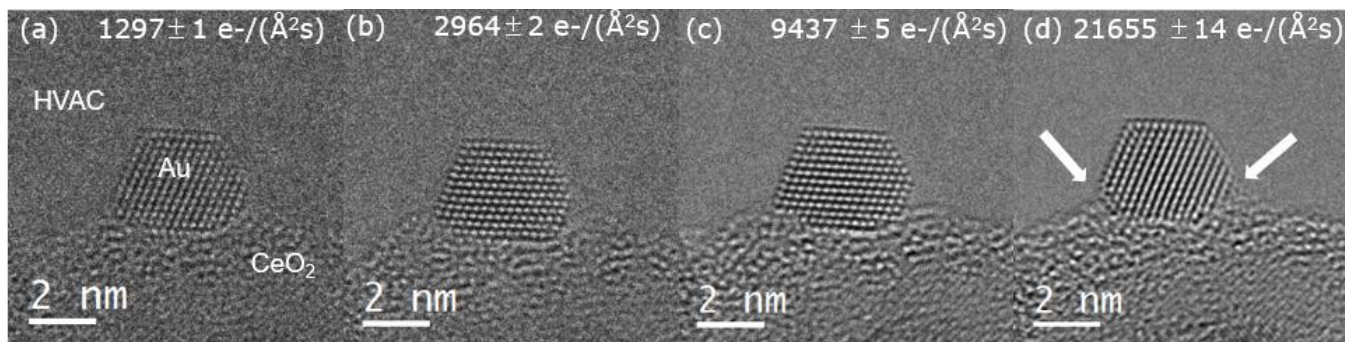


Figure 1. The effect of increasing dose rate. The dose rate shown on each frame is the mean dose rate and standard deviation of the mean. At the highest dose rate, the surface atoms become mobile as indicated by the white arrows.

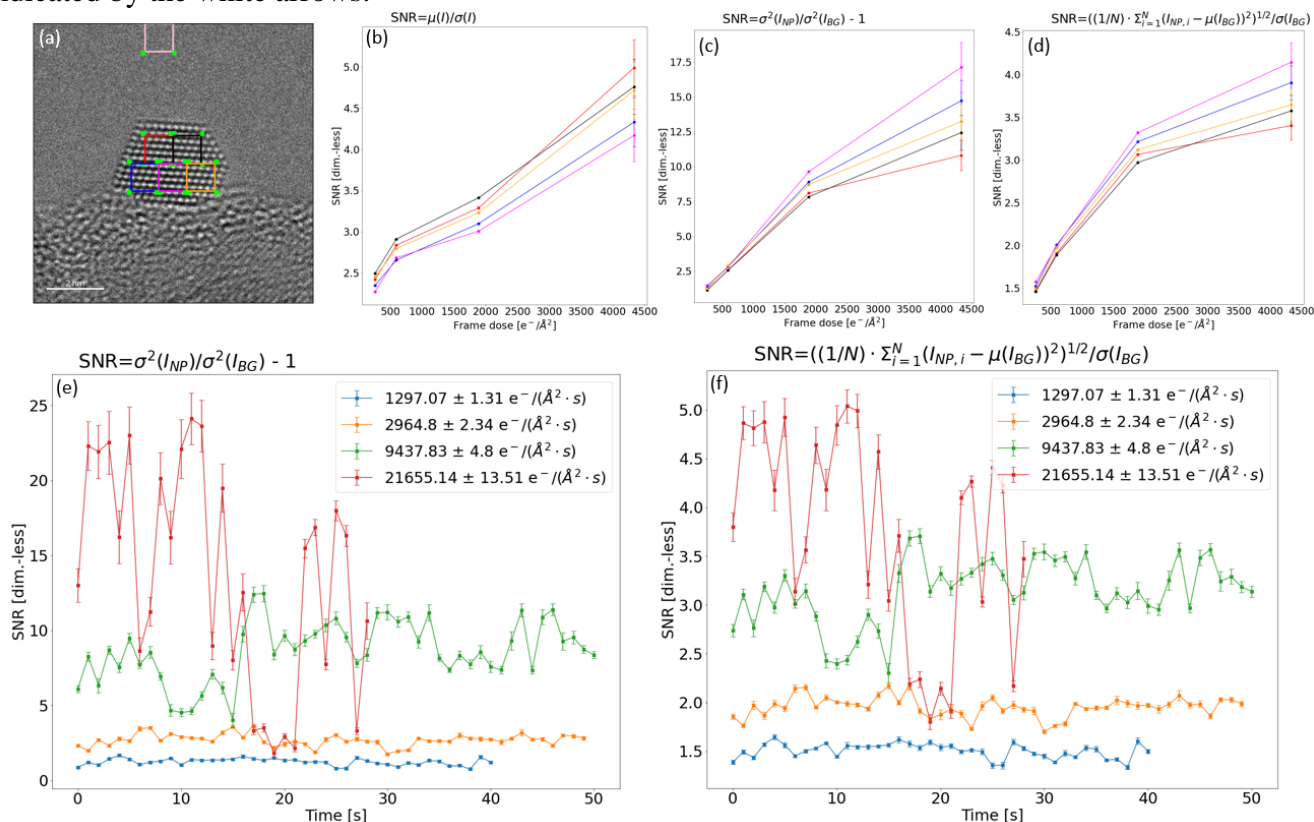


Figure 2. Examples of different models of SNR. (a): Signal measured on different parts of the nanoparticle. The pink square indicates the vacuum measurement. The scale bar is 2 nm. Three different models of SNR: (b): Traditional; (c): model including the variance of the signals; (d): modified model including the mean squared difference between nanoparticle and vacuum. (e)-(f): Temporal variation of the SNR calculated using the models in (c) and (d).

References:

[1] J. Madsen *et al.*, Adv. Theory Simul. **1** (2018), p. 1800037 doi: 10.1002/adts.201800037
 [2] M. De Graef: "Introduction to Conventional Transmission Electron Microscopy", 2003 (Cambridge University Press, Cambridge, United Kingdom)
 [3] A. Rose: "Vision: Human and Electronic", 1973 (Plenum Press, New York-London)

- [4] J. Bushberg *et al.*: "The Essential Physics of Medical Imaging – Third Edition", 2012 (Lippincott Williams & Wilkins, Philadelphia)
- [5] P. Liu *et al.*, *Nanoscale* **11** (2019), p. 11885, doi: 10.1039/c9nr02731a
- [6] P. Liu *et al.*, *J. Phys. Mater.* **3** (2020), p. 024009