The 3D-architecture of individual free silver nanoparticles captured by X-ray scattering

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The diversity of nanoparticle shapes generated by condensation from gaseous matter reflects the fundamental competition between thermodynamic equilibration and the persistence of metastable configurations during growth. In the kinetically limited regime, intermediate geometries that are favoured only in early formation stages can be imprinted in the finally observed ensemble of differently structured specimens. Here we demonstrate that single-shot wide-angle scattering of femtosecond soft X-ray free-electron laser pulses allows three-dimensional characterization of the resulting metastable nanoparticle structures. For individual free silver particles, which can be considered frozen in space for the duration of photon exposure, both shape and orientation are uncovered from measured scattering images. We identify regular shapes, including species with fivefold symmetry and surprisingly large aspect ratio up to particle radii of the order of 100 nm. Our approach includes scattering effects beyond Born’s approximation and is remarkably efficient—opening up new routes in ultrafast nanophysics and free-electron laser science.
Naturally grown particles exhibit a vast variety of architectures ranging from simple, almost spherical shapes (for example, fog droplets), to highly symmetric polyhedral (for example, clusters or certain viruses) and fascinating, complex geometries such as snowflakes or pollen grains. Studying growth processes of nanostructures addresses the key fundamental question of how geometric structure and stability are determined by the occurrence of thermodynamically metastable shapes during particle formation. An illustrative case is the condensation of metal nanoparticles. Albeit optimal equilibrated shapes can be predicted from energetic considerations such as the Wulff construction\(^1\), manifold morphologies are frequently reported\(^2\) that are far away from the ideal equilibrium geometries and often beyond theoretical predictability. Conventional microscopy methods have enabled high-resolution imaging of individual particles grown or deposited on surfaces and revealed insight into their geometric properties\(^3\). Single free, unsupported particles, however, elude experimental access via microscopy since they cannot be immobilized without an interacting substrate. The current knowledge on the morphology of free metal particles is therefore based on orientation- and ensemble-averaged approaches such as electron diffraction\(^6\)–\(^8\), drift measurements\(^9\) and photoelectron spectroscopy\(^10\).

An unambiguous experimental morphology characterization of free particles has to face a number of challenges. First, the co-existence of diverse shapes requires circumvention of ensemble averaging by the study of individual particles. Second, to reliably resolve the particle shape irrespective of its orientation, three-dimensional (3D) imaging methods are required. Third, free particle growth is a statistical process such that a specific combination of shape, orientation and size cannot be repeatedly prepared, excluding tomographic techniques\(^11\)–\(^12\) that rely on multiple measurements of the same object or of equivalent replicas. Considerable efforts have been devoted to obtaining 3D structure information of nanosystems via diffractive imaging with intense femtosecond pulses from X-ray free-electron lasers (X-FELs)\(^13\)–\(^21\). In-flight characterization of single nanoparticles by X-ray small-angle scattering has been successfully demonstrated, revealing the effective two-dimensional (2D) projection of the electron density\(^1\)\(^4\),\(^16\),\(^20\). The 3D reconstruction based on single-shot small-angle X-ray scattering data can only be achieved by exploiting additional symmetry information, as was shown for the case of deposited particles\(^18\). The reconstruction of wide-angle X-FEL scattering has been advocated as an enabling technology for the complete single-shot 3D structure determination of individual nanosystems\(^12\). However, the short X-FEL wavelengths utilized in previous single-shot in-flight particle imaging studies precluded the detection of sufficient wide-angle signal to apply this technology.

Here we show that this limitation can be overcome by employing soft X-FEL pulses. Based on the resulting single-shot wide-angle scattering images, we demonstrate the identification of symmetry, morphology and orientation of individual gas-phase Ag particles by means of a simple and efficient procedure based on fast simulations. Our approach is complementary to the full computational inversion of the scattering process, provided that the particles can adequately be described by a parametric geometry model. The reported results provide evidence for metastable shapes of unsupported Ag particles in a so far inaccessible size regime.

**Results**

**Wide-angle scattering experiment.** The key experimental requirement for the 3D characterization is the ability to resolve the scattering signal up to large angles. For an illustrative motivation of this requirement, it is convenient to assume validity of the first Born approximation. In the limit of small-angle scattering, the scattered far-field can essentially be described by a 2D Fourier transform of the object’s shape projected onto a plane (characterized by normal vector \(\mathbf{n}_p||k_0||\) perpendicular to the incident beam direction, see Fig. 1a. This follows from the fact that the transfer momentum \(q\) is small in magnitude (\(|q| \ll |k_0|\)) and therefore essentially perpendicular to the incident wave vector \(k_0\). The resulting intensity distribution \((i)\) reflects only effective 2D information on the object’s density distribution and (ii) is point-symmetric with respect to \(q = 0\), which impedes unique identification of the target orientation.

These limitations can be overcome by recording scattering under large angles (\(|q| \approx |k_0|\)) because the scattering pattern then reflects \(q\) dependent projections of the density (on planes with normal vectors \(\mathbf{n}_p\parallel|k_0|\)), see Fig. 1b. In a sense, wide-angle scattering enables single-shot tomography as the direction of the projection plane varies with scattering angle within a single image. However, due to the drastic decrease of the scattering intensity \(I(q)\) with increasing \(|q|\) (Porod’s law), significant signal from the particle shape can in practice be detected only up to a critical angle. Considering near spherical shapes, this angle is roughly
Scattering patterns and particle morphologies. The measured single-particle scattering patterns (Fig. 2, left column) show highly symmetric patterns with twofold (a), threefold (b), fivefold (c), and sixfold (d) symmetry. The patterns consist of one or more closed ring-like features near the centre followed by discontinuous higher order rings that form streak-like features accompanied by a faint fine structure. Scattering patterns with odd number of mirror axes (Fig. 2b,c), that is, with broken point symmetry, immediately demonstrate that the wide-angle scattering data contains true 3D structure information. Because of substantial absorption of the soft X-ray radiation inside the particles (penetration depth \( \approx 12.5 \) nm for bulk silver), the morphology identification method must account for scattering effects beyond the Born approximation, which excludes application of conventional iterative reconstruction techniques. We use a simple and efficient multislice Fourier transform (MSFT) algorithm that includes an effective treatment of absorption to calculate scattering images from 3D trial shapes based on a large systematic set of polyhedra (see Methods).

Excellent qualitative agreement between measured scattering images and MSFT results (Fig. 2) is achieved by adjusting size and orientation of the trial model shapes (see Methods for the detailed procedure). In most cases, even fine details are well reproduced, such as the spots in between x-shaped main features in Fig. 2a. Among the morphologies that match the experimental data are decahedra (a), truncated octahedra (b), icosahedra (c) and surprisingly flat hexagonal particles, which correspond to truncated twinned tetrahedra (d). For each of the above shapes, particles with different size and orientation are identified in the data set (see examples in Fig. 2e–h), confirming the repeated occurrence of the identified geometries. Although some of the compact shapes (such as those in Fig. 2b,c) deviate only weakly from a sphere, the scattering images taken for different orientations are extremely diverse (compare Fig. 2b,c to Fig. 2f,g respectively; see Supplementary Movie 1 for a simulation of all high-symmetry orientations of a truncated octahedron). The strong directional and shape sensitivity (even for nearly spherical shapes) demonstrates the 3D capabilities required for the unique identification of particle morphologies. Striking evidence for the transition from small- to wide-angle scattering regimes can be found within a single image for large clusters if it shows broken

![Figure 2](image-url)
point symmetry (see Fig. 2b,g). In these cases, the symmetry changes from even, close to the image centre, to odd in the outer regions.

**Benchmark of the MSFT method.** To corroborate the reliability of the MSFT method and to elucidate the role of absorption, we have compared theory results for different levels of approximation for the case of the truncated octahedron (cf. Fig. 2b). The resulting data within the small-angle approximation (Fig. 3a), implemented via a 2D Fourier transform of the projected scattering density, predicts point symmetric scattering images (the power spectrum of a real-valued function is symmetric) and fails to resemble the experiment. This point symmetry is lifted in the 3D simulation (Fig. 3b) obtained within Born’s approximation. Comparison to MSFT including effective absorption (Fig. 3c) shows that absorption induces a broadening of scattering features, a relative intensity increase in higher diffraction orders and a reduction of the scattering angle of the first-order intensity maximum. Differences between the MSFT result and that of the full treatment of multiple scattering within the finite-difference time-domain (FDTD) framework (Fig. 3c versus Fig. 3d) are more subtle and in most cases irrelevant for shape identification, justifying the effective absorption treatment in MSFT. Compared with the 3D simulations in Fig. 3b–d, the 2D scattering image in Fig. 3a shows similar features close to the centre but deviates significantly at large scattering angles, illustrating the transition from small-angle to wide-angle scattering within a single image.

**Shape refinement.** The MSFT method enables efficient shape identification from a finite set of trial shapes and yields a rough estimate of the parameters of the respective geometry model. A more accurate description of the scattering process via FDTD simulations offers further refinement of the free parameters by direct minimization of the mean-squared deviation of calculated and experimental scattering patterns (see Methods for technical details). This is exemplarily illustrated in Fig. 4 for the truncated octahedron from Fig. 2b, yielding even better agreement between experiment and simulation (R-factor \( \approx 0.18 \), see Fig. 4e). The resulting shape (Fig. 4c) was obtained after optimizing the degree of truncation and the radius of the particle, leading to a slightly reduced value of the predicted radius of \( r = 95 \) nm (Fig. 4d) and a relative truncation of 0.39, as compared with the MSFT estimate of \( r = 100 \) nm obtained under the assumption of an ideal Archimedean shape with a truncation of 1/3. This scheme can be extended to other parameters such as orientation (see Fig. 4e) or shape asymmetries to obtain precise information on the geometry of the individual nanoparticle.

**Discussion**

The variety of particle structures derived from the scattering images in Fig. 2 demonstrates that the motives are much richer than expected from thermodynamic considerations in the investigated size regime. Similar morphologies as those found in this work were reported for preparations involving chemical reactions\(^3\), physically evaporated\(^2\,\text{26} \) or cluster-beam deposited\(^2\,\text{27} \) particles, providing evidence for the general existence of such shapes. The particles in these studies were, however, *supported* by a substrate and are not imaged in the free beam. In the absence of a surface, only truncated octahedra can be derived by free surface energy minimization of particles with face-centred cubic (fcc) lattice structure. Decahedral and icosahedral geometries represent metastable states whose facets cannot be constructed from low-index surfaces of a single fcc crystallite. Particularly striking is the

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**Figure 3 | Comparison of different approximation levels.** False-colour images show the simulated scattering intensity (logarithmic scale) of a truncated octahedron (cf. Fig. 2b) as function of the transverse components of the scattering vector within different approximations. (a) Small-angle approximation corresponding to an effective scattering density projected onto a plane. (b) Born’s approximation taking into account the full 3D geometry but no absorption and refraction. (c) Same as b but including a simplified absorption model. (d) Full FDTD simulations using the optical properties of bulk-silver.
case of unsupported icosahedral particles, which are stable for small clusters but have been predicted to undergo a transition to fcc-derived structures already for sizes as low as a few hundred atoms. Previous observations of icosahedral clusters considerably larger than that (a few 10^6 atoms) have been explained by a non-equilibrium growth process, where isomers being favoured for small sizes provide seed shapes that persist in subsequent stages of particle formation, often resulting in multiple twinned species. The current results demonstrate the existence of free metastable nanoparticles up to much larger radii of ≈150 nm (≈10^9 atoms). The identified highly symmetric metastable shapes reveal that free Ag nanoparticles retain a structural memory of early formation stages up to this so far unexplored size range. This conclusion is corroborated by the observation of strongly oblate particles with aspect ratios of about four (Fig. 2h). The 30–40% larger surface of such shapes substantially increases the surface energy when compared with fcc-derived structures already for sizes as low as a few hundred atoms.

Figure 4 | Optimization of model parameters. (a) Experimental and (b) simulated scattering patterns for a single Ag particle with the optimized shape of a truncated octahedron as depicted in c using the FDTD method (false-colour on logarithmic scale). Parameter optimization for truncation and radius was performed by minimization of the mean-squared deviation (MSD) of experimental data from theory as exemplarily shown in d for the particle size. The optimal radius is \( r = 95 \text{ nm} \) with an uncertainty of ± 8%, estimated from the curvature of the normalized MSD around the minimum. (e) R-factor as a function of rotation angle for the truncated octahedron depicted in e. For these calculations, the model shape is rotated away from the optimal orientation in c around an axis parallel to the upper edge of the hexagonal front facet.

Methods

Particle source. Silver particles were produced by a cluster beam machine equipped with a magnetron sputtering source, operated with Ar and Xe at a pressure of the order of ≈1 mbar. The aggregation section was cooled with liquid nitrogen. No mass selection or ion optics have been used. The clusters were guided into the differentially pumped main chamber through a conical skimmer with 3 mm inner diameter.

Scattering experiment. About 70 cm behind the source, the beam density was low enough to ensure a single nanoparticle at a time in the focal volume (focus size ≈20 μm) of the FEL FLASH at DESY in Hamburg. The power density of the FEL pulses has been estimated from the highest observed charge state of atomic Xe. Typically, Xe^{+11} is found which corresponds to 10^{14}–10^{15} W cm^{-2} at a pulse length of ≈100 fs (refs 33,34). The scattering patterns were recorded shot-to-shot (10 Hz repetition rate) with an imaging detector as described in refs 17,25, covering scattering angles from θ = 3° to 39° over a 2π azimuth. To avoid detector damage from the direct FEL beam, all detector components have a centre hole. The tilt angle of the detector microchannels results in a depletion artefact visible at \((q_x, q_y) ≈ (0.6, -0.4) \text{ nm}^{-1}\) in the scattering images (for example, Fig. 2d). Due to the weak focus conditions (Rayleigh length: some mm; wavelength: 13.5 nm), the
description as a plane wave is justified for the incident beam. In order to maintain single-particle conditions in the focal volume, the hit rate was kept below 10%. Most of the 25,000 scattering patterns with significant signal (from a total of 300,000 recorded images) originated from small clusters resulting in no fringes or from agglomerates of two or more particles (for typical signatures see ref. 35). About 1,000 images were suited for further analysis, for about 100 of them, the particles’ size, shape and orientation have been uniquely identified in this work. The most common symmetric morphologies are truncated octahedra, flat hexagonal shapes, decahedra and icosahedra. Due to manual selection of scattering images for identification, the obtained variety of shapes does not necessarily reflect the actual statistics present in the particle beam.

Experimentally accessible angular range. Within Born’s approximation and under the assumption of free electrons (scattering factors of unity), the scattering cross-section \( \sigma \) of a symmetric polyhedron consists of a central core \( \sigma_{\text{core}} \) (for \( q \ll R_{\text{core}} \)) and an outer envelope \( \sigma_{\text{envelope}} \) (for \( q \gg R_{\text{core}} \)), which is then rotationally symmetric about the incident photon beam. This is in good agreement with the best MSFT estimate. We note that the reduced radius has only minor effects on the scattered far field intensity distributions are obtained using a near-to-far-field transformation of the continuous wave solution. A typical result of the FDTD method is compared with MSFT in Fig. 3c.d.

Refinement of parameters describing the model shape. Refinement of the model shape is done by optimizing additional parameters such as orientation, truncation and size. An upper bound for the error of the parameters specifying the model shape can be estimated via the mean-square deviation of experimental and simulated scattering images. Such a quantitative comparison requires FDTD calculations to accurately include the material properties and knowledge of the detector response as a function of intensity, which is in general nonlinear and may be described by a power behaviour (in our case, the exponent is close to 1/3). The example in Fig. 4 results in a near-parabolic evolution of the deviation as a function of radius, from which the parameter uncertainty that is, the standard deviation of a parameter in a non-linear fit, see ref 39) \( \sigma = 7 \mu m \) can be estimated. This calculation has been performed after optimization of the truncation, hence the radius slightly differs (\( \approx 5 \mu m \)) from the one given in Fig. 2b, where the latter reflects the best MSFT estimate. We note that the reduced radius has only minor effect on the particle volume because of the specific definition of the radius as that of a circumscribed sphere. The factor \( R \) in Eq. 4e was calculated via \( R = \int \left| I_{\text{corr}} - I_{\text{exp}} \right| \frac{d\Omega}{\int \left| I_{\text{corr}} \right| d\Omega} \int \left| I_{\text{corr}} - I_{\text{exp}} \right| \frac{d\Omega}{\int \left| I_{\text{corr}} \right| d\Omega} \), where \( I_{\text{corr}} \) and \( I_{\text{exp}} \) are the experimentally observed and FDTD-calculated intensities in the scattering images, respectively.

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Author contributions

H.H., S.B. and I.B. designed the setup of the cluster source, D.R., L.F., M.S., M.A. and C.B. designed and prepared the scattering setup, D.R., L.F., M.S., I.B., H.H., C.B. and S.S. performed the measurements, R.T. took care of the FLASH laser, C.P. and T.F performed the FDTD calculations, K.-H.M.-B., T.M., D.R., T.F., C.P., H.H. and I.B. analyzed and discussed the results, I.B. performed the MSFT calculations and wrote the manuscript with the input of all co-authors.

Additional information

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