

Electron-Enabled Nanoparticle Diffraction

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We propose a scheme for generating high-mass quantum superposition states of an optically precooled, levitated nanoparticle through electron diffraction at its subnanometer crystal lattice. When a single electron undergoes Bragg diffraction at a free-falling nanoparticle, momentum conservation implies that the superposition of Bragg momenta is imprinted onto the relative coordinate between electron and nanoparticle, which entangles their wave functions. By imaging the electron interferogram, one maps the nanoparticle state onto a superposition of Bragg momenta, as if it was diffracted by its own lattice. This results in a coherent momentum splitting approximately 1000 times greater than what is achievable with two-photon recoils in conventional standing-wave gratings. Self-interference of the nanoparticle can thus be observed within drastically shorter free-fall times in a time-domain Talbot interferometer configuration, significantly relaxing source requirements and alleviating decoherence from environmental factors such as residual gas and thermal radiation. Shorter interference times also allow for a recapture of the nanoparticle within its initial trapping volume, facilitating its reuse in many rapid experimental duty cycles. This opens new possibilities for experimental tests of macroscopic quantum effects within a transmission electron microscope.

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Introduction—Matter-wave interferometry with massive objects challenges our understanding of the quantum-classical transition and is expected to provide necessary experimental insights into quantum gravity [1–9]. State-of-the-art near-field setups have demonstrated interference for particles up to 10^5 amu [10–12] and up to 10^3 amu in the time domain [13]. While standard quantum theory imposes no fundamental limits on particle size, alternative theories suggest a suppression of macroscopic superpositions, calling for experiments that could rule out these theories [14–16]. The use of optically cooled, highly controllable nanoparticles presents a promising platform for such experiments [17–24]. Although numerous proposals have highlighted strategies for high-mass interferometry [3,25–33], experimental realization still remains a challenge and hinges on innovative techniques to realize coherent matter-wave diffraction, reduce environmental disturbances, lower the interference time, and improve experimental stability and repetition rates. While most

approaches rely on optical fields as diffractive elements for matter waves, we propose an electron-enabled nanoparticle diffraction (END) scheme that exploits the underlying atomic structure of a nanoparticle through high-resolution electron imaging. In effect, the particle can be made to diffract off its own crystal lattice, resulting in drastically lower interference times and experimental requirements than other approaches. Table I lists the required free-fall times and distances for nanoparticles of various sizes to interfere, given the exemplary diffraction period of 192 pm.

The END approach takes inspiration from the first matter-wave experiments by Davisson and Germer [34] demonstrating elastic Bragg diffraction of electrons at the atomic lattice structure of a crystalline specimen. If the Bragg condition is met for a given period d of lattice planes, the electron wave function splits into a superposition of diffraction components separated by multiples of the (mass- and velocity-independent) Bragg momentum $p_d = h/d$, where $h = 2\pi\hbar$ denotes Planck's constant. These Bragg components would show up as distinct bright spots on a detection screen in the far field (Fourier plane) or they could be filtered and refocused to form d -periodic interference fringes on a farther image plane. Crucially, momentum conservation dictates that for each Bragg momentum imparted on the electron, the crystal particle receives a recoil of equal magnitude and opposite sign.

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TABLE I. Talbot times and typical free-fall distances for the interference of different nanoparticle masses considered in recent publications [17–22,27] at the exemplary grating period $d = 192$ pm considered here. We require that the particle free falls over twice the Talbot time to observe interference fringes. The short free-fall distances facilitate a recapture of the particle within a single trap volume.

High-mass interferometry $d = 192$ pm			
System	Mass	Talbot time	Free fall
Bateman <i>et al.</i> [27]	1×10^6 amu	9.2×10^{-8} s	1.7×10^{-13} m
Delić <i>et al.</i> [17] (SiO ₂)	2×10^9 amu	1.8×10^{-4} s	6.7×10^{-7} m
Bykov <i>et al.</i> [21] (SiO ₂)	2×10^{10} amu	1.8×10^{-3} s	6.7×10^{-5} m
Jin <i>et al.</i> [22] (diamond)	1×10^{11} amu	9.2×10^{-3} s	1.7×10^{-3} m
Perdriat <i>et al.</i> [18] (diamond)	7×10^{11} amu	6.5×10^{-2} s	8.2×10^{-2} m

In principle, Bragg diffraction in a selected spatial direction can thus generate Einstein-Podolsky-Rosen–like entanglement as it maps product wave functions of electron and particle, $\psi(x)\Psi(X)$, to a superposition of states that are momentum-shifted in the *relative* coordinate $x - X$,

$$\psi(x)\Psi(X) \mapsto \sum_n f_n \exp\left[\frac{i}{\hbar} n p_d (x - X)\right] \psi(x)\Psi(X). \quad (1)$$

By detecting the electron position x on the image plane, where the Bragg orders interfere, one effectively erases which-way information and projects the particle state onto a phase-shifted superposition of Bragg-diffracted components,

$$\Psi_x(X) \propto \sum_n f_n \exp\left[\frac{i}{\hbar} n p_d (x - X)\right] \Psi(X), \quad (2)$$

up to normalization. The outcome looks as if the particle had been diffracted off a grating with a period d given by the particle’s own crystal lattice. Interference between the components can be observed after an additional free evolution time on the order of the Talbot time, $T_M = Md^2/\hbar$, which grows with the particle mass M .

The coherent recoils and their said implications are typically of no relevance in Bragg spectroscopy or in electron imaging of massive (often fixed) crystal samples. However, they can have a sizable effect on levitated, motionally cooled nanoparticles, which have become available only recently in a mass range of $M \sim 10^6$ – 10^{12} amu [17–22]. In fact, the Bragg momenta here are about 1000 times greater than the diffraction momenta of optical grating structures in existing proposals for high-mass interferometers [25–27,30,32], which implies a drastic reduction of the required free evolution times by 1×10^6 for a given mass.

Having said that, the realization of the effective grating transformation (2) requires a precisely timed coherent single-electron pulse to hit the particle and a postselection scheme that filters out all Bragg components other than the multiples of p_d in the electron state. Moreover, since the

superposition in (2) varies in phase with the measured electron position x , the measurement resolution must be better than the angstrom-sized period d . Otherwise, the interference contrast will be reduced. Transmission electron microscopy (TEM) is expected to meet these challenges in the near future; current TEM already enables spatial resolutions < 50 pm [35–38] with highly coherent illumination [39], imaging on the micrometer scale [40], temporal control on the subpicosecond scale [41–43], few- μ rad or better resolution of diffraction angles [44], and near-unitary detection efficiency [45].

Proposed experiment—The proposed experimental sequence illustrated in Fig. 1 adopts the basic interference scheme of Ref. [27], but with significant modifications. The experiment is conducted within an ultrafast transmission electron microscope and utilizes pulsed beams of single electrons that can be imaged with subatomic resolution. In the first stage (a), a nanoparticle is captured and cooled, e.g., in an optical dipole trap [17,46] or ion trap [21,47]. The position of the particle is measured and, e.g., feedback or cavity cooling is applied to further localize the nanoparticle state. The trap thus serves as a pointlike matter-wave source.

After release from the trap, the nanoparticle evolves freely for a time t_0 before (b) a triggered single-electron pulse hits the particle and diffracts off its crystal lattice. The momentum state of the electron thus splits into distinct diffraction components separated by discrete Bragg momenta, each of which imparts an equal momentum recoil on the particle in the opposite direction. A chosen Bragg momentum p_d and multiples thereof are then postselected with the help of a pinhole mask placed in the back focal plane (BFP) of an objective lens (OL), which blocks the undiffracted electron state and any Bragg component other than np_d in a given spatial direction (x axis) [48]. To this end, one must align the particle’s crystal lattice accordingly. The selected Bragg components are allowed to interfere on an image plane further down the beam line, where the electron position x is measured. At perfect resolution, this approximately projects a pure center-of-mass state onto the postmeasurement state (2),

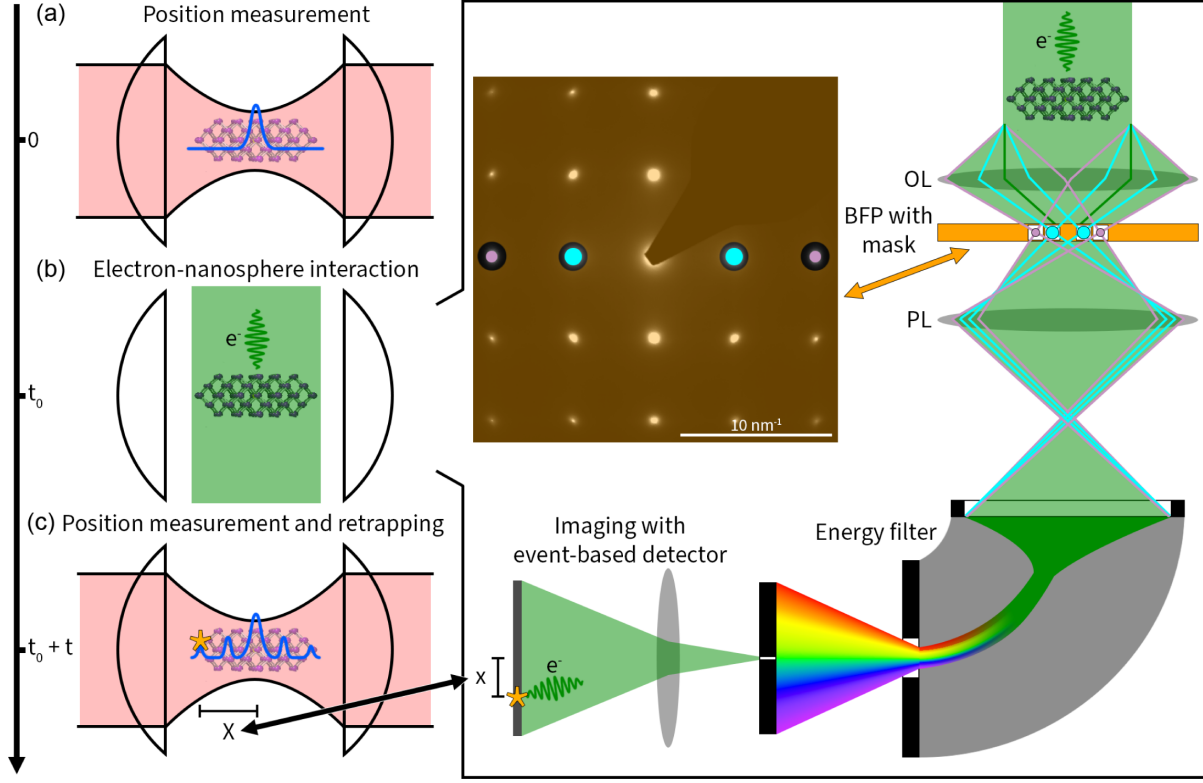


FIG. 1. Proposed scheme of electron-enabled nanoparticle diffraction consisting of three steps: (a) a nanoparticle of mass M is cooled close to its motional ground state in, e.g., an optical trap, acting as a highly localized matter-wave source released into free fall when the trap is switched off. After a sufficient buildup of coherent delocalization over the time t_0 , (b) a triggered single-electron wave packet impinges and Bragg-diffracts off the particle's lattice structure. As each imparted Bragg momentum comes with an equivalent recoil, the wave functions of particle and electron are now entangled. The electron passes a mask on the back focal plane (BFP) of an objective lens (OL), which selects Bragg orders corresponding to a lattice period d . Inset: experimental diffraction image of a silicon crystal, illustrating the intensity distribution when aligned along the relevant zone axis, and the selection of Bragg orders of periodicity $d = 192$ pm. The selected orders are recombined on an image plane by the projector lens (PL) system and an energy filter is used to exclude inelastic events. The detector at the image plane records the electron's position x . This effectively maps the particle wave function into an x -dependent superposition of Bragg momenta. They are allowed to interfere over another free-fall time t , before (c) the position X is measured and the particle retrapped. Given free-fall times of at least one Talbot time, $t, t_0 \gtrsim T_M$, stable interference fringes of magnified period $D = d(1 + t/t_0)$ in the relative coordinate $X - xD/d$ will form over many valid repetitions (i.e., whenever the electron is detected).

equivalent to the effect of a diffraction grating of period d [49]. The nanoparticle can be assumed at rest during this process, which takes a few nanoseconds. Finally, the nanoparticle evolves for another time t before (c) its center-of-mass position X is measured and it is recaptured in the original trap for the next run of the sequence. We discard runs in which the electron is not detected. Over many cycles, the measured position distribution exhibits interference fringes shifted with respect to the electron position x , which can be calculated using standard phase-space methods [49].

Figure 2 depicts exemplary interference fringe patterns we predict assuming perfect measurement resolution. The pattern is given relative to the measured electron position x and would be smeared out accordingly at finite resolution. In our case study, we consider a silicon nanoparticle of mass $M = 2 \times 10^9$ amu and the trap parameters of

Ref. [17] (305 kHz trap frequency, 12 μ K center-of-mass temperature), which results in a Gaussian source state of standard deviation $\sigma_X \approx 3.8$ pm, close to the motional ground state. Our mask shall consist of four pinholes selecting the multiples $n = \pm 1, \pm 2$ of the Bragg order specified by the Miller indices $(1\bar{1}0)$ in primitive-cell representation [i.e., $(20\bar{2})$ in the conventional cubic-cell notation]. The associated grating period is $d = 192$ pm, which demands that the pinholes be a few micrometers in size [48]. To ensure spatial coherence of the nanoparticle over one period, we let it evolve for one Talbot time before the electron interaction, $t_0 = T_M \approx 192$ μ s. In (a), we show the interferogram of the recaptured nanoparticle after varying times t . The fringe period magnifies geometrically as $D = d(1 + t/t_0)$. Notice that the existence of fringes does not certify the quantum wave nature of the nanoparticle. Treating the electron interaction stage as a classical

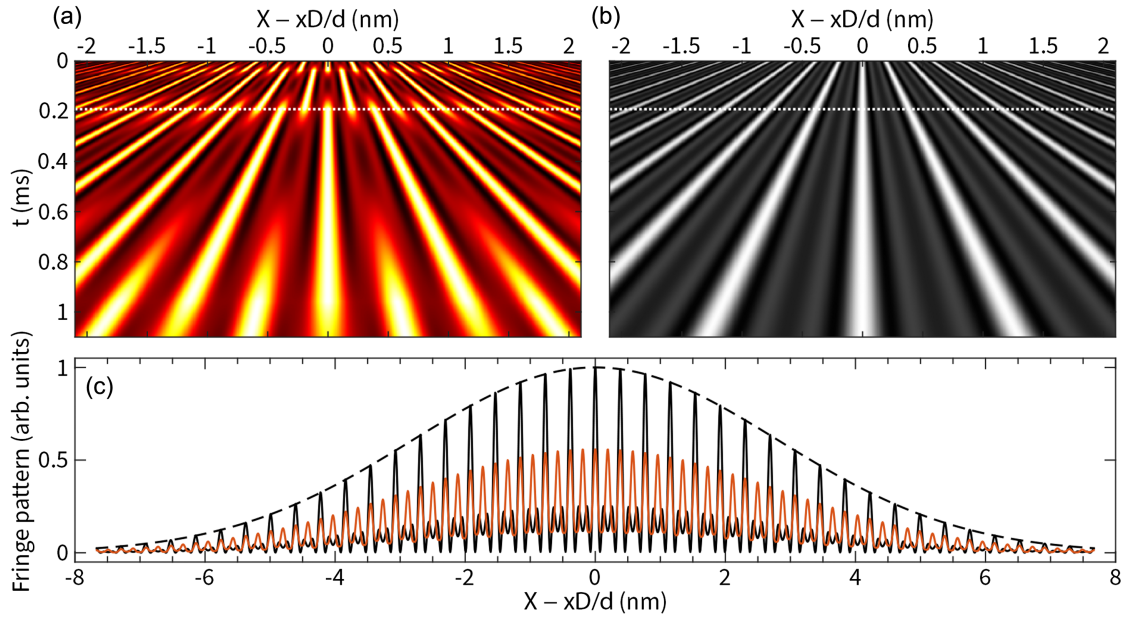


FIG. 2. Exemplary fringe patterns for an aligned silicon nanocrystal of mass $M = 2 \times 10^9$ amu, based on electron Bragg diffraction at $\pm(1\bar{1}0)$, $\pm(2\bar{2}0)$ and plotted against the relative coordinate $X - xD/d$ with respect to the detected electron position x . The particle is released and expands freely for the time $t_0 = T_M = 192$ μ s before the electron diffraction. In (a), we plot the position distribution of the particle as a function of time t after diffraction, normalized to its maximum value at each t . In (b), we show the hypothetical shadow pattern for a classical particle transmitted by a classical aperture. (c) Position distributions at $t = T_M$ [dotted lines in (a) and (b)] corresponding to quantum interference (red solid) and classical shadow fringes (black solid), in units relative to the maximum of the classical pattern. The dashed line indicates the Gaussian distribution of a freely evolved particle without diffraction.

grating aperture would result in the shadow pattern (b), and one cannot claim quantum interference whenever the patterns (a) and (b) coincide. Panel (c) compares the fringe patterns at t_0 , $t = T_M$, where they differ the most. To reveal the quantum fringes, the measurement resolution must be better than $d/4 = 48$ pm for the electron and $D/4 = 96$ pm for the nanoparticle, which are within reach [38,47,52,53]. The short total interference time $t + t_0 = 384$ μ s amounts to a free-fall distance of less than a micrometer, which allows for recapture and thus a fast duty cycle, as we will briefly discuss next.

Our exemplary analysis is carried out for a typical electron energy of 300 keV and a silicon nanoparticle in the shape of an oblate spheroid of radius $R_M = 109$ nm and thickness $2b_M = 60$ nm. This amounts to the mass of a particle for which ground-state cooling was already achieved [17], but our scheme can also be operated with larger particles. The electrons shall have a Gaussian transverse profile with a 115 nm half-width-half-maximum spot size. From this, we estimate the probability to detect the Bragg-filtered electron as $\text{Pr}_{\text{det}} \approx 0.1\%$. Temporal triggering suppresses unwanted detection events due to, e.g., cosmic rays. The duty cycle of our proposed scheme is mainly constrained by the free evolution time, which can be as short as two Talbot times (see Fig. 2). Particles lighter than 2.5×10^9 amu would fall less than 1 μ m and could be easily retrapped within the same potential for reuse in subsequent experiments, allowing for experimental

repetition times of about 1 ms. Consequently, considering Pr_{det} and the Poissonian nature of the electron production process, we estimate approximately 1000 successful experimental runs within 1 h.

Discussion—The proposed electron-enabled nanoparticle diffraction scheme represents a viable route toward high-mass matter-wave experiments with levitated nanoparticles at the mass scale of 10^9 amu and beyond. Leveraging the precise control and high spatiotemporal resolution of present-day transmission electron microscopy, the END scheme exploits the coherent recoil caused by Bragg diffraction off atomic structures, which entangles the wave functions of electron and nanoparticle. Upon electron detection, the particle self-interferes within a short free-fall time of less than a millisecond (at 10^9 amu); see Table I. This not only relaxes the experimental constraints associated with high-vacuum conditions and black-body radiation, but it also facilitates a reliable recapture of the nanoparticles and fast experimental duty cycles.

Our exemplary case study with $M = 2 \times 10^9$ amu and an interference time $t = 1$ ms could reach a logarithmic macroscopicity as high as $\mu \approx 16.3$ —one order of magnitude above the status quo [12,54]. This is based on an empirical measure that compares the “size” of quantum superposition states achieved in experiments with mechanical degrees of freedom [49,55]. Adopting a recent idea for entangling electrons through a quantum scatterer [56], one could envisage whole sequences of consecutive electron

interactions for multiparticle entanglement and interference experiments within shallow nanoparticle traps.

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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End Matter

For the experimental case study discussed in the main text, we make the following assumptions and present estimates about the most relevant systematic effects. We assume that the electrons are only scattered once and elastically, neglecting both multiscattering and inelastic scattering. Multiscattering can either enhance or suppress the amplitudes of certain Bragg peaks, calling for a detailed analysis based on the actual shape and size of the nanoparticle in a realization of our proposal. Given an inelastic mean free path in silicon of about 180 nm [57], unwanted decoherence due to inelastic scattering should be negligible in our case study. To mitigate decoherence for larger particles, the electrons can also be postselected according to the amount of energy they have lost. Modern TEM setups with electron energy filters allow for an energy-filtered imaging resolution of less than 250 meV [58,59].

We further assume that the orientation of the nanoparticle is controlled. This could be achieved, for instance, if the particle exhibits an electric dipole moment and is subjected to a homogeneous electric field; nanoscale

rotational control schemes have been proposed and tested recently [60–62]. The effect of such a static field on the electrons can be compensated using coils that deflect the beam in the opposite direction. If the orientation of the nanoparticle is not controlled, our scheme still works, albeit with a reduced scattering rate into the chosen Bragg peaks and a loss of contrast due to imperfect Bragg peak selection. See Ref. [49] for an exemplary assessment of the visibility loss due to imperfect alignment.

Localized charges attached to the nanoparticle (e.g., remaining surface charges or charges created by the electron beam) will induce an additional electrostatic deflection of the electrons by an angle of the order of $e^2/(2\pi\epsilon_0 m_e \gamma v^2 b)$, where $\gamma = (1 - v^2/c^2)^{-1/2}$ is the Lorentz factor, c is the speed of light, and b is the impact parameter of the electron with respect to the localized charge. For $b = 1$ nm and a kinetic energy of 300 keV, the deflection angle is of the order of 10^{-5} rad and can be safely neglected. Magnetic dipole interactions with unpaired electron spins (e.g., dangling bonds) are even weaker [63].

Another potential risk for the experiment could be the electron-induced sublimation of silicon atoms, changing the mass and potentially the shape of the nanoparticle. However, due to the relatively low electron dose proposed for the successful implementation of the experiment (10^6 electrons per $3.7 \times 10^4 \text{ nm}^2$ cross-section area), the chance of a sputtering event is negligible [64]. Backscattering of a 300 keV electron would impart at most 0.2 mm/s onto the nanoparticle, which does not lead to particle loss.

Optical trapping of nanoparticles can result in high internal temperatures due to photon absorption [65], which may cause a gradual rearranging of atoms in the crystal and thus deformation. This problem could be mitigated by employing a suitable dielectric material with low absorption or by operating with a charged nanoparticle in an ion trap [21,22].

Additionally, incorporating optical or ion traps into the confined space between the pole pieces of a transmission electron microscope is a nontrivial, but feasible, task. The coupling of light into or out of the TEM sample region has been accomplished in many different ways for cathodoluminescence or photon-induced near-field electron microscopy, with the use of fiber optics [66,67] or miniaturized free-space mirrors [68–72] allowing the possibility of free-space diffraction-limited optical imaging of the sample region as well as the measurement of temporal correlations. High intensity laser cavity systems have also been successfully integrated [73], demonstrating the feasibility of establishing strong optical fields inside the transmission electron microscope. Given the experience with such optical systems as well as the growing interest in developing large-gap pole pieces [74–76], we anticipate that it will be possible to incorporate an optical or ion trap into a transmission electron microscope.

Decoherence estimates: Massive neutral particles are subject to decoherence, mainly caused by emission of thermal radiation and by collisions with residual gas particles [77]. The result is an exponential reduction of fringe visibility with an exponent $\Gamma_{\text{dec}}(t + t_0)$, proportional to the total interference time and an effective decoherence rate that depends on the material properties, the mass M , and the fringe period d . Compared to other high-mass interference schemes, the interference time and fringe period are much shorter here, alleviating the impact of decoherence. For a simple conservative estimate, we extrapolate from the experiment proposed in Ref. [27] for a silicon particle of 10^6 amu , with 10^2 ms interference

time, and 10^3 nm fringe period. Here, the mass is more than a factor 10^3 greater, the interference time 10^3 smaller, and the fringe period more than 10^3 smaller. For the case of gas collisions, Γ_{dec} is conservatively estimated by the collision rate, which scales roughly like $M^{2/5}$. Decoherence is therefore *reduced* by about $10^{-9/5}$ compared to Ref. [27]. Decoherence by thermal emission can be modeled as a diffusion process due to the weak recoil of blackbody photons at μm -sized wavelengths. In this case, $\Gamma_{\text{dec}} \propto Md^2$, which yields a decoherence effect suppressed by 10^{-6} relative to Ref. [27]. Hence, decoherence should be irrelevant for vacuum pressures of 10^{-9} mbar , which is achievable with modern TEM [78], and internal particle temperatures below their melting point.

Dephasing due to charging of the nanoparticle can also be safely neglected. The dipole trap is placed in between the pole pieces of the transmission electron microscope, providing a spherical volume with a radius $r = 2 \text{ mm}$ of vacuum to decouple the nanoparticle from the environment. Charging of the nanoparticle with an additional electron will cause an electrostatic deflection due to mirror charges of $\Delta s = (e^2/4\pi\epsilon_0 r^2)t^2/2M$, which amounts to $\Delta s \approx 1 \text{ pm}$ within two Talbot times, much smaller than the fringe spacing d .

Spatial resolution: An important technical requirement for the proposed experiment is to resolve the interference fringes of the electron and the nanoparticle. On the electron side, our proposal demands a spatial resolution of about $d/4 = 48 \text{ pm}$. The imaging resolution is mainly limited by lens aberrations or system instabilities [35]. Chromatic aberrations can be minimized using chromatic aberration correctors [79,80], or by reducing the energy spread present in the image via incident beam monochromation [81] or postselection energy filtering [82]. Recent progress in the correction of spherical aberrations resulted in a third-order aberration coefficient as small as $C_s = 350 \text{ nm}$, which theoretically supports a resolution down to $d_s = \lambda(C_s/2\lambda)^{1/4} = 34 \text{ pm}$ [35]. In practice, the resolution is currently still limited by higher-order aberrations to about 50 pm , which will continue to improve in ongoing efforts.

On the nanoparticle side, one must be able to resolve distances $D/4 = 96 \text{ pm}$ upon recapture. State-of-the-art cavity-enhanced schemes allow position measurements at the $10^{-14} \text{ m}/\sqrt{\text{Hz}}$ level [52], while more conventional setups have demonstrated a position sensitivity below 100 pm within less than $20 \mu\text{s}$ [83].