# Synthesis of high-purity solid $SiO_2$ nanodumbbells via induced aggregation for levitated optomechanics

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## Abstract

Optically levitated nanodumbbells in vacuum are excellent candidates for thermodynamics, macroscopic quantum mechanics, precision measurements and quantum sensing. Silica ($SiO_2$) material, with extremely low absorption of near-infrared light and super mechanical strength, has been the most potential material for optically levitated systems. Here we synthesize high-purity solid $SiO_2$ nanodumbbells via Stöber method by introducing acetone for the induced aggregation of $SiO_2$ nanospheres. The nanodumbbells show high uniformity and their sizes are tunable. Previous experimental results demonstrated that the synthetic nanodumbbells can be applied in GHz nanomechanical rotors and can withstand the tensile strength of over 13 GPa. This work supports batch production and high yield of $SiO_2$ nanodumbbells, which engineers a new material platform to advance levitated optomechanics.

**Keywords:** Silica ($SiO_2$), nanodumbbell, Stöber method, induced aggregation, levitated optomechanics

## Introduction

Levitated optomechanics provide a powerful platform for nonlinear dynamics, precision sensing, rotational quantum mechanics and multiple other applications [1-5]. Optically levitated spherical particles have been applied for force sensing at the level of $10^{-21}$ N [6] and for interaction detecting of dark energy [7]. Recently, optically levitated nonspherical nanoparticles were proposed and showed superior performance than spherical particles in record-high GHz nanomechanical rotor [8, 9], Casimir torque measurement [10], rotational matter-wave interferometers [11], and high-frequency gravitational wave detection [12]. However, the large centrifugal force in high-speed rotation and the damage of photothermal effect require super tensile strength and extremely low absorption of light of the material.

Silica ($SiO_2$) nanoparticles, owing to their excellent biocompatibility, thermal stability, facile synthetic route, and batch synthetic availability, have been widely applied in various fields such as biotechnology, energy, electronic, sensor and catalysis [13-20]. Some methods for the preparation of silica nanoparticles are employed such as chemical vapor condensation, micro emulsion processing, combustion synthesis and sol-gel processing [21-25]. $SiO_2$ nanodumbbells, with nonspherical geometry, low absorption of visible and near-infrared light and robust mechanical strength, have been the most promising material in the field of optically levitated nonspherical systems. Although the nanodumbbells have been synthesized by microemulsion method [26], wherein surfactant molecules driving two nanospheres in a micelle and the nanodumbbell was formed under depletion effects induced aggregation [27, 28]. However, surfactants that used to form microemulsions introduce light-absorbing impurities into the $SiO_2$ and weaken the mechanical strength of the material [29-31], resulting in the disassembling of nanodumbbells in our previous optically levitated experiments [8]. Stöber method, which does not use long-chain surfactants, is the most widely used wet chemistry synthetic approach to high-purity solid monodisperse $SiO_2$ nanospheres [32-34]. For the growth of nanodumbbells, induced aggregation of two nanospheres is the most important step. Ammonia can be used to aggregate micro-sized $SiO_2$ particles, but it is ineffective on $SiO_2$ nanospheres [26]. Therefore, it is of great significance to develop a method that can effectively aggregate $SiO_2$ nanospheres and achieve batch production of high-purity solid nanodumbbells.

Herein, by introducing acetone for the induced aggregation of $SiO_2$ nanospheres, we successfully synthesize the solid $SiO_2$ nanodumbbells via Stöber method. With a circularly polarized laser, the nanodumbbell can be driven to rotate beyond 1 GHz in high vacuum, which is the fastest nanomechanical rotor realized to date [8]. Remarkably, the ultimate tensile strength of the synthetic nanodumbbells exceeds 13 GPa, which is 2 orders larger than the bulk glass.

## Experimental Section

**Schematic Design.** Fig. 1 shows the schematic illustration of the $SiO_2$ nanodumbbells synthetic processes. Solid $SiO_2$ nanospheres are synthesized via Stöber process (Fig. 1A), then acetone is added into the solution to induce aggregation of nanospheres (Fig. 1B). Next, a small amount of TEOS is added to grow silica shells under stirring (Fig. 1C). Finally, monodisperse nanodumbbells are obtained after the removal of nanospheres and other conglomerates by differential velocity centrifugation (Fig. 1D).

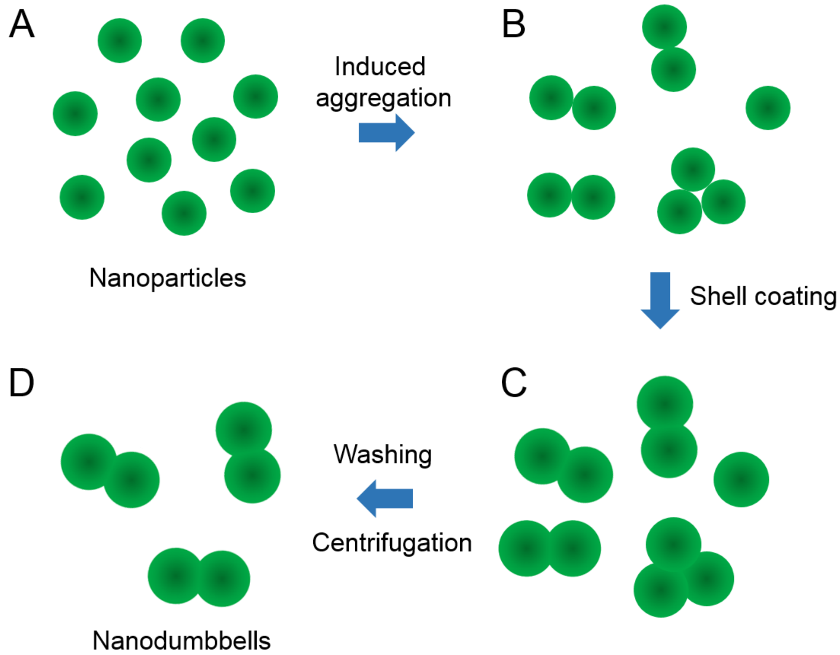


**Fig. 1** Schematic illustration of $SiO_2$ nanodumbbells synthetic processes. (A) Solid $SiO_2$ nanospheres are synthesized via Stöber method. (B) Induced aggregation of the nanospheres by acetone. (C) Growing silica shells to enhance the mechanical strength of nanodumbbells. (D) Monodisperse nanodumbbells are obtained after the removal of nanospheres and other conglomerates.

**Chemicals.** All reagents are of the highest purity possible to avoid the introduction of impurities resulting in optical absorption. All lab glasswares are soaked in aqua regia for 24 hours and flushed with deionized water for 3 times. Tetraethyl orthosilicate (TEOS, 99.999%) is purchased from Aladdin reagent. Ammonia (26.5% w/w of $NH_3$ in $H_2O$), acetone (>99.9%) and ethanol (>99.9%) are all chromatographic grade and purchased from Sinopharm Chemical Reagent Ltd., China. Deionized water with a resistivity of 18.2 MΩ·cm (Milli-Q) is used for all rinsing processes. All reagents are used as received without further purification.

**Nanodumbbell Synthesis.** $SiO_2$ nanospheres with a diameter (d) of 80 nm are synthesized by adding 2 mL of TEOS to a well-stirred mixture of ammonia (9.72 mL), deionized water (5.96 mL) and ethanol (200 mL). After stirring for 48 h, a colloidal solution of $SiO_2$ nanospheres with a diameter of about 80 nm is obtained. Then 20 mL of acetone is added into the colloidal solution and stirred for 24 h to induce aggregation, and 40 μL of TEOS is added under stirring for another 24 h to grow the silica shells. The silica shell can enable the nanodumbbells to have larger ultimate tensile strength [35, 36]. The precipitate of silica nanodumbbells with a diameter (d) of 90 nm is obtained after washing with ethanol and centrifugation at 9000 r.p.m. for 10 minutes.

**Purification of Nanodumbbells.** For the requirement of optical levitation in high vacuum, the $SiO_2$ nanodumbbells must be high-purity to avoid the damage of photothermal effect. So the product is washed by anhydrous ethanol for seven times, and then transferred into a Teflon-lined stainless-steel autoclave (50 mL capacity) and heated at 90 °C for 12 h. Subsequently, the removal of nanospheres and other conglomerates will be carried out by differential velocity centrifugation [37-39]. After centrifuge the product at 1000 r.p.m. for 10 minutes, the supernatant is collected and centrifuged at 9000 r.p.m. for another 10 minutes. Furthermore, the product is centrifuged at 3000 r.p.m. for 5 minutes, the supernatant is collected and centrifuged at 6000 r.p.m. for another 5 minutes. Finally, the precipitate is dried under vacuum at room temperature overnight and the purified monodisperse $SiO_2$ nanodumbbells are produced.

**Characterization.** Scanning electron microscopy (SEM) images are obtained using a Helios NanoLab DualBeam electron microscope with an accelerating voltage 5 kV. Energy dispersive X-ray spectrum (EDX) is also characterized in the SEM with an accelerating voltage of 10 kV. Transmission electron microscopy (TEM), high-resolution transmission electron microscopy (HRTEM) and selected area electron diffraction (SAED) images are acquired using Tecnai F30 microscope operated at 300kV.

## Results and discussion

Based on above processes, high-purity solid $SiO_2$ nanodumbbells are successfully mass-produced. Fig. 2A shows the monodisperse solid $SiO_2$ nanospheres synthesized by Stöber method. Since $SiO_2$ nanospheres has a lower solubility in acetone than in ethanol, acetone can be applied to induce the aggregation of $SiO_2$ nanoparticles. Fig. 2B shows the aggregated nanoparticles induced by acetone, meanwhile the nanospheres, nanodumbbells and other conglomerates are coexisting and their surfaces are not covered with shells. In order to further enhance the mechanical strength of nanodumbbells so that they will not be dismembered by large centrifugal force in high-speed rotation, extra silica shells are grown on the previous aggregated nanoparticles (Fig. 2C). The diameter (d) of silica cores will also be a little bit larger after coating. Finally, after purification by washing and differential velocity centrifugation, monodisperse solid $SiO_2$ nanodumbbells are obtained (Fig. 2D).

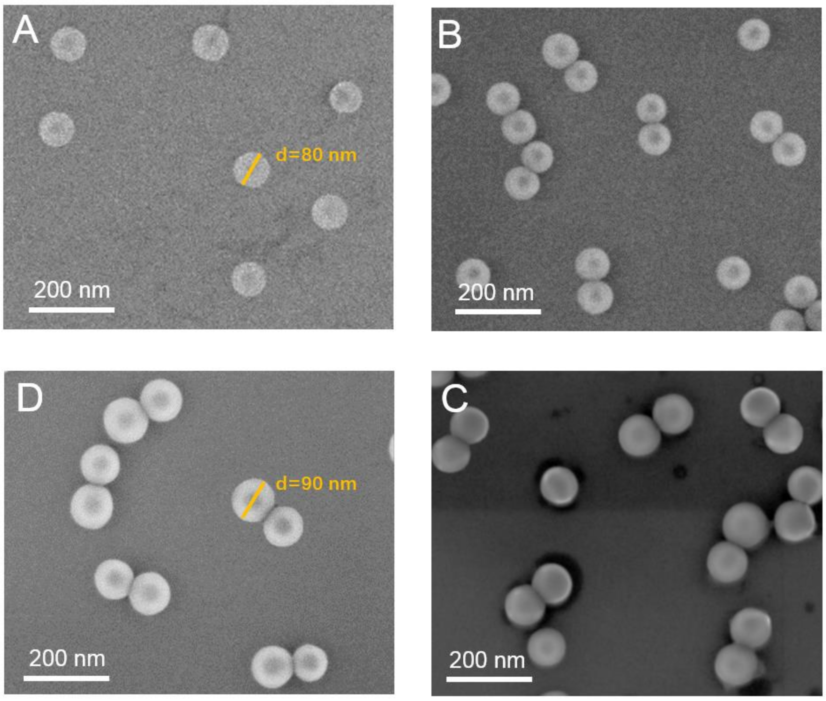


**Fig. 2** SEM images of various growth stages. (A) monodisperse solid $SiO_2$ nanospheres. (B) The aggregated nanoparticles induced by acetone. (C) Extra silica shells are grown on the previous aggregated nanoparticles. (D) High-purity solid $SiO_2$ nanodumbbells.

Fig. 3A shows the TEM, HRTEM and SAED characterizations of $SiO_2$ nanodumbbells. As shown in the HRTEM image, the structure of $SiO_2$ is solid rather than mesoporous. The solid structure ensures the robust mechanical strength due to the high density of covalent bonding in $SiO_2$. SAED characterization confirms the amorphous structure of $SiO_2$. It has shown that the ultimate tensile strength of the synthetic nanodumbbells is 2 orders larger than the bulk glass [8, 40]. The explanation for the increased tensile strength observed in the $SiO_2$ nanodumbbells is the reduced possibility of having large cracks in nanodumbbells than the bulk glass [40]. EDX analysis of $SiO_2$ nanodumbbells shown in Fig. 3B. The only existing elemental composition of Si and O determined from the EDX spectrum indicate the high purity of nanodumbbells and prove that there is no acetone or ethanol molecules trapped in nanodumbbells.

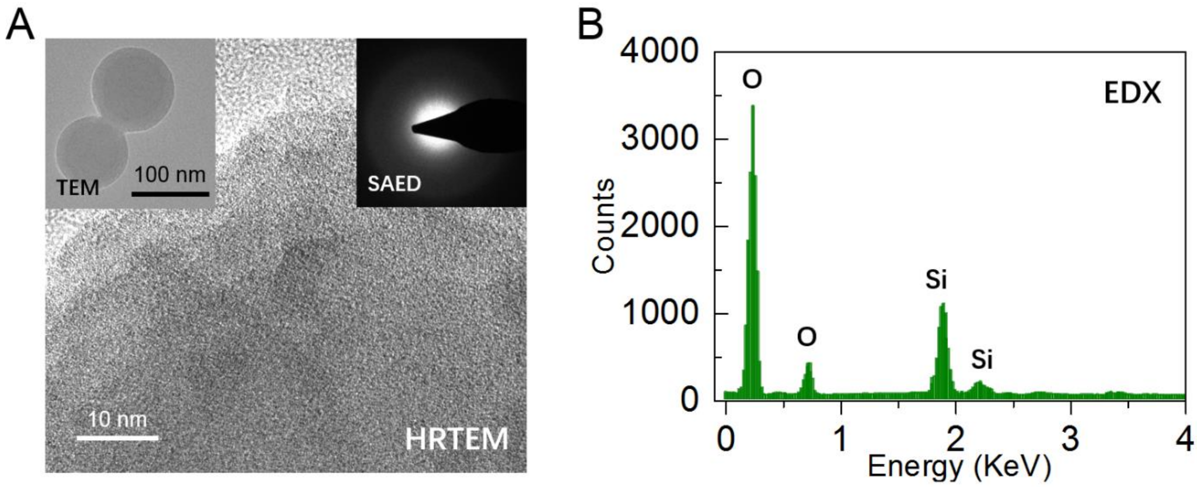


**Fig. 3** Structure and purity analysis of nanodumbbells. (A) TEM, HRTEM and SAED images of $SiO_2$ nanodumbbells. The structure of nanodumbbells is solid and amorphous. (B) EDX analysis of $SiO_2$ nanodumbbells. The only existing elemental composition of Si and O determined from the EDX spectrum indicate the high purity of nanodumbbells and prove that there is no acetone or ethanol molecules trapped in nanodumbbells.

The diameter of nanodumbbells is tunable via our method. As shown in Fig. 4, we also synthesize various nanodumbbells with a diameter (d) of 160 nm (Fig. 4A) and 200 nm (Fig.

4B) by aggregating nanospheres with different nanoparticle sizes. Remarkably, the calculated result proves that the torque detection sensitivity of nanodumbbells under optical levitation would increase as the size of the nanodumbbells decrease [8]. The higher detection sensitivity of the nanorotor will be more conducive to quantum sensing and precision measurements. Stöber method supports batch production and high yield of $SiO_2$ nanodumbbells, but the process could be a bit complex. We also develop a simple physical method to assemble nanodumbbells, but the drawback is that not every particle would be a nanodumbbell, and the tensile strength is not exactly consistent between each nanodumbbell [8]. Nevertheless, nanodumbbell which synthesized by physical synthesis is equally to be used for optical trapping in vacuum after multiple selection.

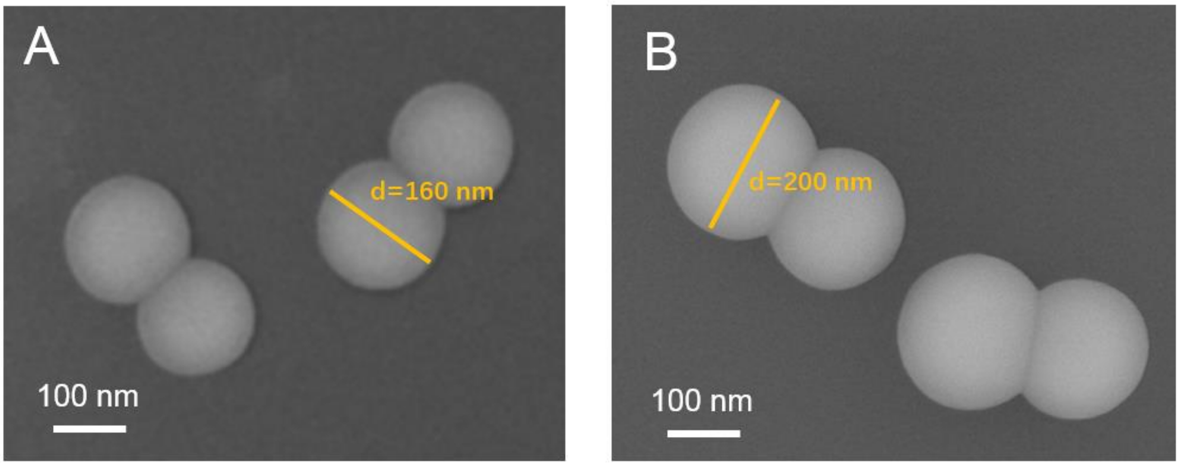


**Fig. 4** SEM images of size-tunable nanodumbbells. Various nanodumbbells with a diameter of 160 nm (A) and 200 nm (B). These nanodumbbells are synthesized by similar process and aggregating nanospheres with different nanoparticle sizes.

## Conclusions

Herein, batch production of high-purity solid $SiO_2$ nanodumbbells are synthesized via Stöber method by introducing acetone for the induced aggregation of $SiO_2$ nanospheres. By avoiding the long-chain surfactant, the synthetic nanodumbbells remain solid structure and extremely low absorption of light. Experimental results show that our $SiO_2$ nanodumbbells can be driven to rotate beyond 1 GHz with a circularly polarized laser in high vacuum, the ultimate tensile strength of the synthetic nanodumbbells exceeds 13 GPa. This approach provides an excellent candidate material in optically levitated field for quantum sensing, precision measurements and other applications.

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**Contributions：** All design, experiments, data analysis and paper writing in this paper have done by Yu-Hao Deng.

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