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## Sensors and Actuators A: Physical



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#### ARTICLE INFO

Article history: Received 7 July 2011 Received in revised form 30 January 2012 Accepted 31 January 2012 Available online 9 February 2012

Keywords: Nano-particles Dielectrophoretic Microspotting Pearl chain formation Temperature Impinging jet

#### ABSTRACT

We show that pearl chains of gold nano-particles (Au NPs) are formed consistently between microelectrodes by combining microspotting and dielectrophoretic (DEP) techniques. Experimental results on various sized Au particles and DEP parameters, including voltage and frequency, are reported in this paper to explore the critical parameters in controlling the pearl chain formation (PCF) process between microelectrodes. PCF is observed from 10 kHz to 5 MHz for 100 nm diameter Au NPs, and 100 kHz to 10 MHz for 10 nm Au NPs. Variations in formation rate are detected when the applied voltage and particle size varies. At higher voltages, PCF occurs at a higher rate and the formation time decreases. The optimum frequency for Au NPs PCF shifts to a higher frequency region when the particle size decreases. Theoretical analysis is carried out by calculating the DEP force with AC electrokinetics to explain the observations at DEP frequencies ranging from 10 Hz to 10 MHz. Finally, Au NP chains formed between the microelectrodes are shown to vary in resistance consistent with predictions for a simplified model of an impinging jet system, indicating that these Au particle sensors could potentially be used to precisely measure localized temperatures and other localized thermal phenomena.

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#### 1. Introduction

Gold nanoparticles (NPs) or colloids have been extensively studied because of their potential applications in nano-medicine [1,2], nano-photonics [3] and nano-devices [4]. In the past few decades, various nanowire fabrication techniques were developed. One common method of nanowire growth is the vapor–liquid–solid (VLS) synthesis method, as first reported in 1964 by Wanger and Ellis [5]. Semiconductor nanowires are fabricated via using gold NPs as catalysts. Recently, a dielectrophoretic (DEP) technique is used to manipulate gold NPs directly [2,6].

The DEP technique assembles nanowires in a single step [7], in which dielectric particles experience a force enhanced by a nonuniform electric field as a result of polarization. However, during

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DEP manipulation, a large power density is generated in the fluid surrounding the electrodes when a high electric field is applied to manipulate small particles [9]. In order to achieve the optimum formation rate for a gold (Au) pearl chain, both theoretical and experimental studies are presented in this paper. Moreover, to minimize the production time and cost, we have combined the DEP gold pearl chain batch manipulation and micro-spotting technology. As a result, Au NP-based sensors can be precisely and effectively fabricated.

#### 2. Theory

#### 2.1. DEP manipulation of nanoparticles

DEP is defined as the lateral motion generated on uncharged particles due to polarization induced by non-uniform electric fields [7]. The ensemble averaged DEP force exerted on NPs over time can be written as:

$$\langle F_{\text{DEP}}(t) \rangle = 2\pi \varepsilon_m a^3 \operatorname{Re}[K(\omega)] \nabla \left| E_{\text{rms}} \right|^2$$
 (1)

where  $\omega$  is the angular frequency,  $|E_{rms}|$  is the time-averaged value of the root-mean-square (rms) of the electric field intensity,  $\varepsilon_m$  is the dielectric permittivity of the medium, *a* is the radius of the

<sup>☆</sup> The preliminary results for this paper have been presented at the 3rd IEEE International Conference on Nano/Micro Engineered and Molecular Systems (IEEE NEMS 2008; ISBN: 978-1-4244-1907-4; Digital Object Identifier: 10.1109/NEMS.2008.4484496).

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<sup>0924-4247/\$ -</sup> see front matter © 2012 Elsevier B.V. All rights reserved. doi:10.1016/j.sna.2012.01.046



**Fig. 1.** The real part of the CM factor for gold colloidal particles with 10 nm and 100 nm diameters suspended in deionized water.

particles, and *K* is the Clausius-Mossotti (CM) factor which is given by:

$$K(\omega) = \frac{\varepsilon_p^* - \varepsilon_m^*}{\varepsilon_p^* - 2\varepsilon_m^*} \tag{2}$$

where  $\varepsilon_p^*$  and  $\varepsilon_m^*$  are the complex permittivity of the particle and the medium, respectively. The complex permittivity is given by:

$$\varepsilon^* = \varepsilon - j\frac{\sigma}{\omega} \tag{3}$$

where  $\sigma$  is the conductivity of the medium [9].

The magnitude and direction of the DEP force are governed by the magnitudes of the applied electric field intensity, the freqency, phyisical dimensions of the NPs, and the real part of the CM factor, respectively. The electric field intensity is determined by both the applied voltage and the geometry of the electrodes. The former factor is proportional to the magnitude of the DEP force and the latter factor determines the formation location of the NP chains.

The frequency of the input alternating current (AC) signal governs the CM factor. The real part of the CM factor determines the direction of the DEP force and it varies from -0.5 to 1.0 for spherical particles. For a positive CM factor, a positive DEP force will be generated. NPs will move towards a region with a strong electric field and hence are drawn downward to the edges of the electrodes. The opposite situation occurs for a negative DEP [2]. Fig. 1 shows the real part of the CM factor for single-shell sperical particles. In our experiment, gold colloidal particles are coasted with a chemical stabilizer; therefore, the particles are considered as a single shell model instead of a homogeneous dielectric sphere [11].

In general, NP chain formation appears to initiate at the location with the highest electric field with the corresponding highest field gradient intensities. Usually, when a NP, under a positive DEP force, reaches the electrode, it will have the same potential as the electrode and thus the electric field intensity becomes higher at its free end. Other NPs in the bulk fluid will then migrate towards the free end (the high electric field region). Eventually, a chain of NPs will form across the electrode gap.

#### 2.2. Electrically induced fluid flow

In our experiment with manipulation of NPs, NPs are dispersed inside a liquid medium. Fluid flow, which is induced by an electrothermal body force and AC electroomosis, is utilized in the manipulation process. An electrothermal force is induced because of a high power density is generated in the fluid surrounding the



**Fig. 2.** A diagram showing the definition of the parameters  $\theta$  and r in Eq. (4).

microelectrode when a high electric field is used to manipulate small particles [8]. The averaged electrothermal body force on the fluid can be expressed as

$$\langle f_e \rangle = \frac{2}{\pi^3 k} \frac{\varepsilon \sigma V_{\rm rms}^4}{r^3} \prod \left( 1 - \frac{2\theta}{\pi} \right) \hat{\theta}$$
 (4)

where the two different  $\theta$  angles are defined in Fig. 2 and

$$\prod = \left[\frac{\alpha - \beta}{1 + (\omega \tau_q)^2} - \frac{\alpha}{2}\right]$$
(5)

where  $V_{\rm rms}$  is the rms of the applied potential, k is thermal conductivity of the medium,  $\alpha = (1/\varepsilon)(\partial \varepsilon/\partial T)$ ,  $\beta = (1/\sigma)(\partial \sigma/\partial T)$  and  $\tau_q$  is the charge relaxation time  $(\varepsilon/\sigma)$ . An order of magnitude estimate for a typical fluid flow velocity can be calculated by:

$$|u| = \left| f_e \right| \frac{l_o^2}{\eta} \tag{6}$$

where  $l_0$  is the characteristic distance of the microelectrode [9].

A simulation estimating the order of magnitude of the maximum fluid flow velocity ( $\theta$  equal to 0 or  $\pi$ ) under different medium conductivities is shown in Table 1. The magnitude of the induced fluid flow velocity is proportional to the conductivities of the mediums. The applied field frequency determines the direction of the flow. When the frequency of the applied signal is smaller than the relaxation frequency, fluid flows away from the inter-electrode gap and competes with the DEP manipulation. The opposite situation occurs when the applied signal is higher than the relaxation frequency.

In addition to the fluid flow generated by the electrothermal force, fluid flow is also induced by an AC electroomosis effect in the low field frequency range. This happens since the charges in a layer between the surface and the electrolyte, called the electric double layer, generates a force when an electric field is applied tangentially to a surface bathed in an electrolyte. Consequently, the movement of charges in the double layer pulls the fluid along the surface and generates a flow which is opposite to the direction of the DEP manipulation [9]. The fluid flow velocity strongly depends on both the frequency of the applied electric field and the conductivity of solution.

The double layer is not only generating the AC electroomosis effect, but also decreases the magnitude of DEP force. The double layer induced by the electrostatic potential attracts ions of opposite charge from the solution and repels ions with same charge. A thin layer near the electrode surface is densely packed with counter ions and loosely packed with co-ions. Eventually, the applied potential drops across the double layer and the actual potential passing through the electrolyte decreases. Thus, the magnitude of the DEP force for manipulating the NPs decreases. This dropping ratio is strongly dependent on the frequency of the applied field and the conductivities of the medium [9,12].

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