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ORIGINAL ARTICLE

Contributions to Plasma Physics

Quantum dot on a plasma-facing microparticle surface: Requirement on thermal contact for optical charge measurement

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Abstract

Semiconductor nanocrystals, quantum dots (QDs), are known to exhibit the quantum-confined Stark effect which reveals itself in the shift of their photoluminescence spectra in response to the external electric field. It was, therefore, proposed to use QDs deposited on the microparticle surface for the optical measurement of the charge acquired by the microparticles in low-temperature plasmas. Another physical process leading to the shift of the photoluminescence spectra of the QDs is heating. Charging of plasma-facing surfaces is always accompanied by their heating. Thermal balance of a quantum dot residing on the surface of a microparticle immersed in a plasma is considered in this work. It is shown that under periodically pulsed plasma conditions, the spectral shift of the photoluminescence of the quantum dot caused by the oscillations of its temperature becomes undetectable if the effective thermal flux characterizing the thermal contact between the quantum dot and the microparticle exceeds the value ranging from 10^8 to 10^{10} s^{-1} depending on the plasma parameters. If this effective thermal flux exceeds the above-mentioned values, the entire spectral shift observed during the period of plasma pulsing should be attributed to the quantum-confined Stark effect due to the microparticle charge. Lower-boundary estimate for the effective thermal flux for the direct contact between the quantum dot and the microparticle is $\sim 10^{12}$ s^{-1}. Estimations based on the heat conductivity of ligand-stabilized nanocrystal arrays yield even higher values of the effective thermal flux $\sim 10^{13}$ s^{-1}.

KEYWORDS

dusty plasmas, optical charge measurements, quantum dots

1 | INTRODUCTION

In-situ measurement of the charge accumulated on small particles is a problem in many areas of science and technology, for example, in the investigations of dust-growing reactive plasmas,[1–3] in the usage of complex (dusty) plasmas as atomistic model systems for the investigations of classical condensed matter phenomena,[4,5] in the investigations of Lunar

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environment,[6] in the usage of microparticles as small optically manipulated plasma probes[7] as well as in the investigations of triboelectric charging.[8–10] Traditionally, the charge of dust particles is measured using dynamical methods[11–16] which have known disadvantages such as necessity for (often not easily verifiable) assumptions on the forces acting on dust particles and limited spatiotemporal resolution.

Optical methods of dust particle charge measurement are therefore of great interest. Significant theoretical efforts have been undertaken to explore the possibilities of optical detection of the dust charge. The surplus electrons modify the dielectric permittivity of the dust particle material or surface conductivity of the microparticle and therefore affect some of the spectral features of light scattering. Possibilities of using the transverse optical phonon resonance[17,18] were considered. Very recently, the possibility of the measurement of charges of silica nanoparticles in plasmas using Fourier-transform infrared spectroscopy of this resonance was experimentally demonstrated.[19]

In addition, very recently, it was shown that semiconductor nanocrystals, quantum dots (QDs),[20–22] deposited on a large flat plasma facing surface are sensitive to the charge on this surface[23] due to the quantum-confined Stark effect[24]: Spectrum of their photoluminescence experiences red shift in reaction to the local electric field. It was therefore proposed[25] to design a charge microsensor by attaching the QDs to the surface of a micrometre-sized spherical particle. The calculations showed that under typical conditions of dusty plasma experiments, such a microsensor can exhibit Stark shifts of the order of fractions of a nanometre. The advantage of using the QDs compared to excitonic resonance is that the measurement can be performed in the visible spectral range.

Experiments[23] have shown that exposure of QDs to the fluxes of charged particles is connected not only with the surplus-charge-induced Stark shift, but also with the thermal shift of the photoluminescence spectrum.[26–28] Charged particles bring their kinetic as well as potential energy to the plasma-facing surface, which is cooled by neutral gas as well as by thermal radiation.[29–32] Heating is, therefore, along with charging, an unavoidable consequence of the exposure of the surface to any ionized medium. The spectral shifts caused by these two phenomena have to be therefore distinguished.

Charging of a plasma-facing surface usually occurs much faster than heating. On this basis, the experimentally observed 'fast' red shift[23] was attributed to the Stark effect. In accord with that, periodic pulsing of the plasma (on the timescales between those of charging and heating) was suggested[25] to distinguish between the thermal and electrostatic effects on the QD photoluminescence spectrum. The weak point of this approach is that this is, in fact, not the surface temperature, but rather the temperature of the QDs which determines the thermal spectral shift. QDs on the plasma-facing surface would not necessarily acquire the same equilibrium temperature as the substrate surface itself. In addition, their thermal inertia is much less than that of the substrate they are sitting on. These two issues raise the question about the thermal contact between the QD and the surface it is attached to: Is it sufficient to suppose the QD being thermally bound to the surface?

This is the question we are addressing in the present paper. Analysing the thermal balance of the QD residing at the surface of a micrometre-sized particle, we arrive to the quantitative requirements for the thermal contact between the QD and the microparticle. We compare the obtained results with the estimations of the thermal contact between QD and the microparticle in case of the QD residing on the microparticle surface without any chemical bonding and in case of the QD attached to the microparticle surface with the help of chemical ligands.

2 | MODEL

We model the thermal balance for both the microparticle and the QD residing on a microparticle surface considering the heat fluxes from the plasma to their surfaces and vice versa as well as heat exchange between them. The fluxes included into the model are schematically shown in Figure 1. Along with earlier works,[29,30,33] we suppose that energy is brought to plasma-facing surfaces by charged species and is taken away by neutral gas and thermal radiation.

If the plasma is periodically pulsed, it periodically goes through reignition and afterglow phases, which are very hard to properly model. Afterglow is quite a long process.[34] Even milliseconds after switching off the electrical power, significant densities of the charged species remain in the reactor volume. Therefore, in a real afterglow, the microparticle and QD surfaces will continue collecting heat. On the other hand, after the plasma reignition, the densities of the charged species and, consequently, the heat fluxes to the microparticle and QD surfaces also do not immediately reach the steady-state values. Exact calculation of the heat balance of a microparticle and a QD residing on a surface of a microparticle under these transient conditions is impossible. To simplify the situation, we suppose that (a) the charged species in the plasma acquire the steady-state densities immediately after the plasma electrical power is switched on and (b) the density of

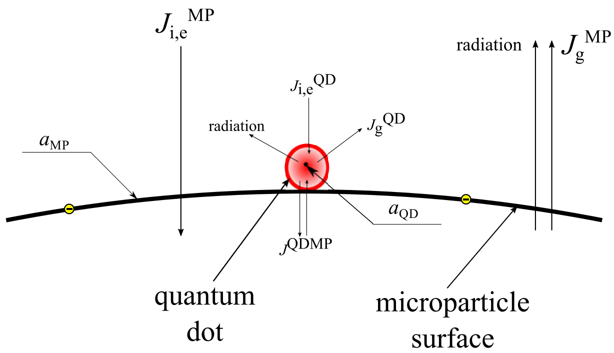

FIGURE 1 Schematic representation of the thermal balance for a plasma-facing microparticle surface and quantum dot residing on it. The surfaces are heated by the fluxes of ions and elecrons $J_{i,e}^{MP,QD}$ and cooled by the fluxes of neutral atoms $J_g^{MP,QD}$ and thermal radiation. Flux J^{QDMP} characterizes the thermal contact between the microparticle surface and quantum dot

the charged species in the plasma drops to zero immediately after the plasma electrical power is switched off. Since the plasma pulsing period should be much longer than the microparticle charging time[25], we additionally assume that (c) the microparticle acquires the steady-state electrostatic potential immediately after the plasma electrical power is switched on and (d) discharges to zero electrostatic potential immediately after the plasma electrical power is switched off. Then, the heat fluxes from the plasma to the surfaces will also bounce between their steady-state values in the reignition and zero in afterglow. This simplification will increase the thermal contrast between the reignition and the afterglow and, therefore, strengthen the requirement to the thermal contact.

The frequency of plasma pulsing should be selected in such a way that the oscillation of the temperature of the microparticle is negligibly small. Then, we are only interested in the variations of the QD temperature which will, in the worst case, oscillate between the temperature of a microparticle (in the afterglow) and the QD temperature in the steady-state plasma (in the reignition). The thermal balance equations for the former and the latter temperatures can then be, respectively, written as

$$\eta\left(J_i^{MP}\left[E_{ion}+e\phi_{MP}\right]+J_e^{MP}k_BT_e\right)-\xi_{MP}J_g^{MP}k_B\left(T_{MP}-T_g\right)-4\pi a_{MP}^2\varepsilon_{MP}\sigma\left(T_{MP}^4-T_g^4\right)=0, \tag{1}$$

$$J_i^{QD}\left(E_{ion}+e\phi_{QD}\right)+J_e^{QD}k_BT_e-\xi_{QD}J_g^{QD}k_B\left(T_{QD}-T_g\right)-4\pi a_{MP}^2\varepsilon_{QD}\sigma\left(T_{QD}^4-T_g^4\right)$$
$$-J^{QDMP}k_B\left(T_{QD}-T_{MP}\right)=0, \tag{2}$$

where η is the duty cycle of plasma pulsing, $J_{e,i,g}^{MP,QD}$ are the electron, ion and neutral gas fluxes to the microparticle and QD surface, respectively, $T_{e,g,MP,QD}$ are the electron, neutral gas, microparticle, and QD temperatures, respectively, $a_{MP,QD}$ are the radii of the microparticle and QD, respectively, $\varepsilon_{MP,QD}$ are the thermal emissivities of the microparticle and QD, respectively, $\phi_{MP,QD}$ are the electrostatic potentials of the microparticle and QD, respectively, $\xi_{MP,QD}$ are the coefficients of thermal accommodation of the neutral gas atoms on the microparticle and QD surfaces, respectively, E_{ion} is the ionization potential of the plasma gas atoms and J^{QDMP} is the effective heat flux characterizing the thermal contact between the microparticle and the QD which we will for simplicity term ‘thermal contact flux.’

First, we consider the isotropic fluxes to the microparticle surface. For the fluxes of charged particles, we follow the approach in which the orbit-motion-limited current is implied for electrons, whereas for ions, the collisions with neutrals in the vicinity of the microparticle have to be considered.[35] The electron flux is then expressed as follows:

$$J_e^{MP}=\pi a_{MP}^2 n v_{eth}\exp\left(-\frac{e\phi_{MP}}{k_BT_e}\right), \tag{3}$$

where $v_{eth}=\sqrt{8k_BT_e/\pi m_e}$ is the electron thermal velocity and ϕ_{MP} is the absolute value of microparticle surface potential (which is normally negative). The expression for the flux of cold (ion temperature equals T_g) ions reads:

$$J_i^{MP}=\pi a_{MP}^2 n v_{ith}\left(1+\frac{e\phi_{MP}}{k_BT_g}+\frac{R^3}{a_{MP}^2 l_i}\right), \tag{4}$$

where R denotes the radius of a sphere around a microparticle, inside which the absolute value of the electrostatic potential is below T_g, $v_{ith} = \sqrt{8k_BT_g/\pi M}$ (M is the ion mass) is the ion thermal velocity and $l_i = k_BT_g/p\sigma_{ig}$ (σ_{ig} is the cross-section of ion-neutral collisions) is the ion mean free path. In case of Yukawa potential around the microparticle, R should satisfy the equation:

$$\frac{e\phi_{MP}}{k_BT_g}\exp\left(-\frac{R}{\lambda_D}\right) = \frac{R}{a_{MP}}, \tag{5}$$

where $\lambda_D = \sqrt{\varepsilon_0 k_BT_g/ne^2}$ is the Debye length.

For the neutral flux onto the microparticle, we use the kinetic expression[30]

$$J_g^{MP} = 2\pi a_{MP}^2 v_{gth}\frac{p}{k_BT_g}, \tag{6}$$

where p is the neutral gas pressure and $v_{gth} \equiv v_{ith}$ is the thermal velocity of neutral atoms.

The microparticle potentials ϕ_{MP} is determined by the balance of electron and ion fluxes: $J_i^{MP} = J_e^{MP}$. The charge is then connected to the potential by the expression $Z_{MP}e = 4\pi\varepsilon_0 a_{MP}\phi_{MP}$. It was shown[25] that typical distance between the electrons on the microparticle surface $l = \sqrt{4\pi a_{MP}^2/Z_{MP}} \ll a_{QD}$. Therefore, we can assume that the QD will most of the time stay uncharged and $\phi_{QD} = \phi_{MP}$. In addition, for the microparticle, the fluxes of the plasma species are isotropically collected over its entire surface area, which will not be true for the QD. The respective fluxes will then read:

$$J_{e,i,g}^{QD} = \alpha_{e,i,g}J_{e,i,g}^{MP}\left(\frac{a_{QD}}{a_{MP}}\right)^2, \tag{7}$$

where $\alpha_{e,i,g}$ is the fraction of the total QD surface area over which the electron, ion and neutral fluxes are respectively collected.

Obviously, at arbitrary "surface" coefficients $\varepsilon_{MP,QD}$, $\xi_{MP,QD}$ and $\alpha_{e,i,g}$, temperatures $T_{MP,QD}$ will be different. Our goal is to find such value of J^{QDMP} that $|T_{QD} - T_{MP}| < \delta T$, where δT is the temperature difference corresponding to the spectral shift detection threshold which was estimated as 0.02 nm. The thermal red shift of the CdSe QD photoluminescence is ~ 0.1 nm K^{-1}[26–28] which corresponds to $\delta T = 0.2$ K.

Since the surface coefficients are, in general, unknown, we cannot directly solve the Equations (1) and (2). We, therefore, undertake further simplifications. We note that usually $E_{ion} > k_BT_e$. The ions will therefore be the main heat carrier in our system. To significantly decrease the temperature difference between the microparticle and the QD, the heat exchange between these bodies has to dominate over other cooling mechanisms. Therefore, if $J_i^{QD} \lesssim J^{QDMP}k_B\delta T$, the temperature difference between the microparticle and the QD will be below the required threshold. This reduces our uncertainty to only one coefficient α_i which can be constrained to lie between 1/4 and unity. Since α_i cannot be very small, we can assume $J_i^{QD} \sim J_i^{MP}(a_{QD}/a_{MP})^2$ which allows us to define the threshold thermal contact flux

$$J_{thr}^{QDMP} = J_i^{MP}\frac{E_{ion} + e\phi_{MP}}{k_B\delta T}\frac{a_{QD}^2}{a_{MP}^2}, \tag{8}$$

so that for $J^{QDMP} \gtrsim J_{thr}^{QDMP}$, $|T_{QD} - T_{MP}| < \delta T$.

3 | RESULTS

Calculations are performed for the parameters listed in Table 1. The microparticle size and electron temperature were taken from a previously published work[15] (these values were also used in our previous work[25]), where the microparticle charge was measured using the phonon spectra of a 2D plasma crystal. We note that in that case, the microparticle levitated in the sheath of a rf discharge and had, therefore, about factor of three larger charge than that given by the condition $J_i^{MP} = J_e^{MP}$. In addition, to comply with our previous work,[25] we took $a_{QD} = 3.3$ nm. Plasma density n was varied between 10^{14} and 10^{16} m^{-3}, whereas the gas pressure p were varied from 0.4 to 40 Pa. This range of experimental conditions covers almost the entire parameter space available in the capacitively-coupled rf discharge which is often used in the dusty plasma experiments.

TABLE 1 Input parameters for the calculations of the threshold flux J_{thr}^{QDMP}

Parameter	Value
$k_B T_e$ (eV)	1.3
T_g (K)	300
δT (K)	0.2
M (a.m.u.)	40
σ_{ig} (m^{-2})	10^{-18}
E_{ion} (eV)	15.76
a_{MP} (μm)	4.6
a_{QD} (nm)	3.3

Note: The plasma is supposed to be produced in argon gas.

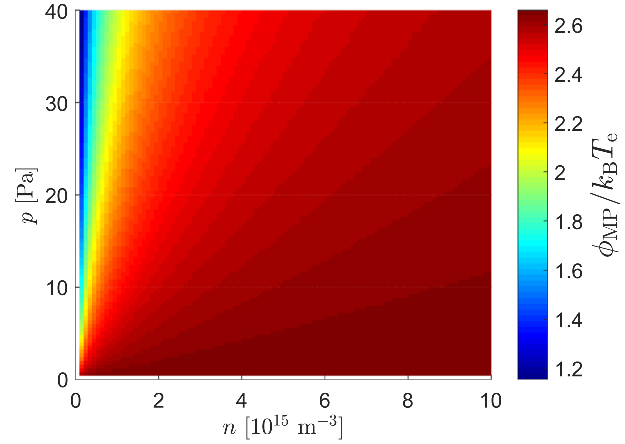

FIGURE 2 Plasma density and pressure dependence of the steady-state elctrostatic potential of the microparticle calculated from the balance of electron (Equation 3) and ion (Equation 4) fluxes

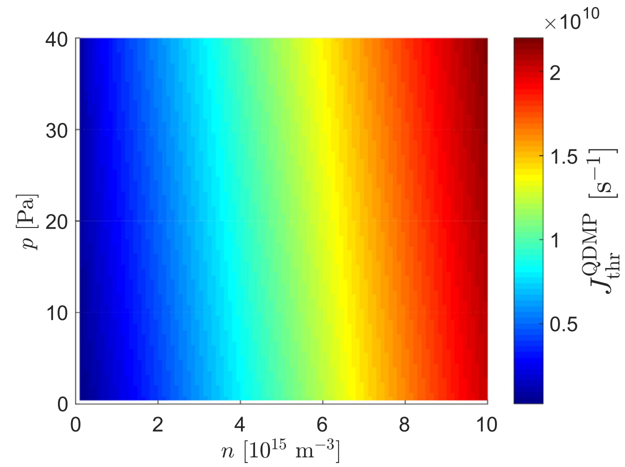

FIGURE 3 Plasma density and pressure dependence of the threshold thermal contact flux J_{thr}^{QDMP} calculated from Equation (8). For effective flux characterizing the thermal contact between the microparticle and QD $J^{QDMP} \gtrsim J_{thr}^{QDMP}$, the temperature difference between the microparticle and the QD during the plasma pulsing will always lie below the detection threshold $\delta T = 0.2$ K

Figure 2 shows the pressure and plasma density dependence of the steady-state electrostatic potential of the microparticle. Both pressure and plasma density variations are associated with the collisional term in Equation (4): Ion mean free path decreases with pressure causing the growth of collisionality and, consequently, the decrease of ϕ_{MP}, and Debye length decreases with plasma density causing the decrease of the ion trapping radius R and, consequently, increase of ϕ_{MP}.

In Figure 3, the plasma density and pressure dependence of the threshold thermal contact flux is shown. The main trend is the linear growth with plasma density, whereas a much weaker increasing trend with pressure due to the increase of the collisional term in Equation (4) is also visible. In the considered parameter range, $J_{\mathrm{thr}}^{\mathrm{QDMP}}$ varies between 10^8 and 10^{10} s^{-1}.

4 | DISCUSSION AND CONCLUSION

For simplicity, let us first assume that the thermal contact between the QD and the microparticle is provided by a pair of atoms one of which belongs to the QD and the other belongs to the microparticle. This approach may be considered relevant for the cases when the QDs reside on the substrate without any chemical bonding.[23] In this case, $J^{\mathrm{QDMP}} \sim \sqrt{\mu_1/\mu_2}\nu_{\mathrm{E}}$, where $\mu_1/\mu_2 < 1$ is the ratio of the average masses of the atoms of the microparticle and QD materials, and ν_{E} is the Einstein frequency of the hotter material. Supposing, in accord with previous works,[23,25] that the QD consists of CdSe, the Einstein temperature T_{E} if of the order of 100 K.[36] Frequency $\nu_{\mathrm{E}} = k_{\mathrm{B}}T_{\mathrm{E}}/h$ and $J^{\mathrm{QDMP}} \sim 10^{12}$ s^{-1} which significantly exceeds the $J_{\mathrm{thr}}^{\mathrm{QDMP}}$ values in Figure 3.

Measurements of heat conductivity of CdSe nanocrystal arrays stabilized by oleic acid[37] which is one of the ligands used to attach the CdSe quantum dots to silica surfaces[38] yield the heat conductivity $\chi \approx 0.2$ W $\mathrm{K}^{-1}\mathrm{m}^{-1}$. Thermal contact flux $J^{\mathrm{QDMP}} \sim \chi h_{\mathrm{l}}/k_{\mathrm{B}}$, where h_{l} is the ligand length. Assuming $h_{\mathrm{l}} \sim 1$ nm for oleic acid, we arrive at $J^{\mathrm{QDMP}} \sim 10^{13}$ s^{-1} which again substantially exceeds the required minimum of $J_{\mathrm{thr}}^{\mathrm{QDMP}}$.

We have therefore shown that the oscillations of the temperature of the QD residing on the microparticle surface exposed to pulsed plasmas are very unlikely to produce a measurable influence on the QD photoluminescence spectrum. This statement applies to the wide (two orders of magnitude) range of plasma density and gas pressure. Other parameters, for example, electron temperature or microparticle size, can be varied in much narrower boundaries and their effect is therefore much weaker.

The possibility of the measurement of the microparticle charges in plasmas using the quantum-confined Stark effect on the QDs attached to the microparticle surfaces has already been demonstrated in our previous work.[25] In that work, we have suggested to use pulsed plasmas for the microparticle charge measurement to exclude the effect of heating of the microparticles in plasmas. By excluding the thermal issue in the present work, we can state that the entire red shift of the QD photoluminescence observed in pulsed plasmas should be attributed to the quantum-confined Stark effect originating from the surplus electrons residing on the microparticle. The proposed measurement technique can therefore be tested experimentally.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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