THE PHYSICS OF MACRO-, MICRO- AND NANOMATERIALS

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Abstract. *Interest in micro- and nanomaterials comes from the fact that matter at the micro- and nanoscale behaves differently from our common macroscopic world. Effects negligible at the macroscopic level become important at the micrometer and nanometer scales, and vice versa. In this communication, the variations of some physical effects with dimensions are discussed, in order to understand the origin of the apparent different properties of materials and their processing. The limits of validity of well known physical laws are also discussed. Particular emphasis is given to nanomaterials, in order to determine when and how size and quantum effects play major roles.*

1. Introduction

There is currently a growing interest in the field of nanotechnologies, both from the fundamental and applied points of view. Nearly 40 countries throughout the World have decided that nanosciences and nanotechnologies are among their scientific and technological priorities. Simultaneously, the interest in micrometer sized components continue to grow and lead to various applications, although some fundamental questions remain.

Interest in micro- and nanomaterials comes from the fact that matter at the micro- and nanoscales behaves differently from our common macroscopic world. Effects negligible at the macroscopic level become important at the micrometer and nanometer scales, and vice versa.

When going from the macro- to the nanoworld, one passes through two boundaries. When the characteristic dimensions of the elements decrease from the macroscopic to the micrometer size, the effects of gravity become negligible as compared with adhesive and friction effects. For instance, surface tension dominates gravity. This implies that our reasoning, based on our experience at the macroscopic level, is no more valid. We have to modify our rules of thinking. The boundary between the macroscopic and microscopic levels is not sharp. It depends on the effect to be considered.

When the characteristic size decreases further to attain the nanometer range, another limit is encountered. While the macroscopic properties of matter remain generally valid at the micrometer size, surface effects become dominant at the nanometer scale. Moreover, when one reaches the interatomic distance range, quantum effects appear.

The problem is further complicated by the fact that:

1) these two boundaries are not sharp ;
2) they depend on the effect being studied;

3) they depend on the nature of the material(s) involved.

Another aspect makes the reasoning disturbing: some effects are quantitatively well described by theoretical models which are not valid (from the fundamental scientific point of view) at the involved size ranges. The Occam'razor principle, or law of parsimony, often applies in nanosystems. One example (among others) is the well known classical nucleation theory. This theory assumes that the concept of surface tension remains valid, while the nuclei are so small (they contain a few hundreds of atoms) that the concept is no more valid. But it works well, and the nucleation theory is used by scientists and engineers.

In order to understand the transition from macroscopic to nanoscopic dimensions, one obviously needs to consider pluridisciplinary effects. In order to go progressively, the starting point would be to deal with scaling laws. In this communication, it is not possible to be exhaustive. So, we choose to treat a few particular cases and restrict our analysis to the "usual" physics at small velocities, where the relativistic effects do not come into play. The limits of validity of some well known physical laws are discussed.

2. From macro- to micromaterials

When going from the macroworld to micron sized materials, the intrinsic properties of the material itself remain generally valid. However, when dealing with materials processing, care has to be taken, due to the facts that :
   a) surface tension effects are much larger than gravity below, say the mm range;
   b) the dimensions of the particles compare with critical transport characteristic lengths, like mean free paths.

Let us consider elements with a typical linear dimension, L. In the following, except when explicitly stated, *it is assumed that all the linear dimensions vary proportionally to L*. This implies that areas, S, vary like L^2 ; volumes, V, vary like L^3. Some scaling laws are given in *Table 1*. As seen from the table, among all the physical parameters, some behave differently with L. For instance, this is the case of F_{gr} and F_{vdw}. Their relative values also vary with L. The adhesion force dominates the gravitational force at low L. The critical value at which both forces are equal depend on the nature of the medium between the two solids. However, below L 1 mm [1], F_{gr} is much less than F_{vdw}. Gravitation may then be neglected at such small dimensions, both in the micro- and the nano-worlds.

Let us now consider a few particular examples.

2.1. Flow in micropipes

Fluids are obviously important, since, except in vacuum, all bodies move in a fluid (air, water,...). Materials processing also often involve fluids. It is then interesting to look at the variation with L of various fluid parameters.

When a body falls « *vertically* » in a fluid, viscous friction is such that, after some time, it falls with constant velocity, v_{lim}. The transient time, τ, is proportional to v_{lim}. If the body is spherical, with radius r, the limiting velocity is given by : $v_{lim} = 4\rho g r^3/18\eta r$, where η is the dynamic viscosity of the fluid and ρ is the density. Viscous forces then drastically and rapidly damp any motion at small dimension. A very small body remains immobile in air (when there

is no air movement). When air moves, the small body follows the air movement, as experienced by us by looking at dust in sunrays.

At high velocities, the hydrodynamics is unstable, and turbulent flow arises. The transition from laminar to turbulent flow is given by the Reynolds number, Re. Re = $\rho v L/\eta$. If $v \sim L$, then Re $\sim L^2$. For flow in pipes, the transition from laminar to turbulent flow occurs when Re $\cong 10^3$. Moreover, it seems that the value of Re at the laminar-turbulent transition diminishes when L is less than about 100 µm [2]. In this case, turbulence disappears in microsystems in which liquids flow. For instance, let us take a capillary tube with diameter : $L = 50$ µm. The velocity of the fluid is $v = 0.5 \times 10^{-3}$ m·s⁻¹ (or $v = 10.L$. s⁻¹). It yields that in air, Re = 0.0016. In water, Re = 0.025. The flows are then laminar and viscous forces are dominant.

The fact that viscous forces are dominant implies that motion is more difficult at small size than could be expected from our (macro)experience. For instance, although the kinetic rotation energy is very low at high angular rotation speed, it is difficult to attain very high rates, due to viscosity effects. Water acts more like a syrup at these small scales.

2.2. Gas flow

When dealing with gas flow in micropipes, one attains regimes where the mean free path of the particles or heat diffusion lengths are of the order of the dimensions of the pipes. The gas flow is described by the kinetic theory of gases. The mean free path of molecules, λ, is given by $\lambda = kT/(4\pi R^2 p)$, where R is the radius of the molecule, p is the pressure... For air, at $T = 288$ K, $\lambda = 6{,}2 \times 10^{-5}$ /p m (p in hPa).

The flow characteristics may be described by means of the so-called Knudsen number, Kn :

$$\text{Kn} = \lambda/L,$$

where L is a characteristic dimension of the pipe (in a cylindrical pipe, L is its diameter).

When a gas flows in a cylindrical pipe, one distinguishes three flow regimes [3] :
1) viscous when $p \cdot L > 0{,}7 \times 10^{-2}$ hPa·m, i.e. $L > 100 \cdot \lambda$ or Kn $< 10^{-2}$;
2) Knudsen when $2 \cdot 10^{-4} < p \cdot L < 0{,}7 \cdot 10^{-2}$ hPa.m, or $10^{-2} <$ Kn < 0.3 ;
3) molecular when $p \cdot L < 2 \times 10^{-4}$ hPa·m, i.e. $L < 3 \cdot \lambda$, or Kn > 0.3.

In the viscous flow regime, the conductance of the pipe (the ratio between the gas flux and the variation of pressure between the two borders) is proportional to p, while, in the molecular flow regime, it does not depend on p. When L decreases, one goes rapidly from the viscous to the molecular regime. For air, at room temperature and atmospheric pressure, the molecular regime is attained at $L < 186$ nm. When $L = 1$ µm, p = 0,2 atm; when $L = 1$ cm, p = $0{,}2 \cdot 10^{-4}$ atm.

These few examples show that the materials behave differently at the micron than at the macrosizes. Other examples are given elsewhere [4, 5].

Table 1 : *Scaling laws* [4,5]

Physical quantity	Definition	Scaling laws	Remarks
Mass	m	L^3	
Gravitation force	$F_{gr} = mg$	L^3	
Pressure on the ground	$p_{gr} = F_{gr}/S$	L	
Adhesive force	F_{vdw}	L^2	Van der Waals, Casimir
Macroscopic friction force	$F_{fr} = \mu F_{gr} = \mu mg$	L^3	
Microscopic striction force	F_{str}	L^2	
Kinetic energy	$E_c = mv^2/2$	L^3	v constant
		$L^5 \ (v \sim L)$	$v \sim L$
Gravitic potential energy	$E_{pot} = mgh$	L^3	h constant
		L^4	$h \sim L$
Moment of inertia	$I = cst.mL^2$	L^5	
Kinetic energy of rotation	$K = (1/2)I\omega^2$	L^5	ω constant
Maximal deflection of a bent beam	ζ	L^2	Under own weight
Fundamental oscillation frequency	ν	L^{-1}	Tubes, beams, cables,….
Maximum velocity of a spherical mass in a fluid	$v_{lim} = 4\rho gr^3/18\eta r$	L^2	
Reynolds number	$Re = \rho vL/\eta$	L	v constant, ρ constant
		L^2	$v \sim L$, ρ constant
Diffusion time	$\tau_{diff} = L^2/\alpha D$	L^2	D constant
Electrical resistance	$R_{el} = \rho_{el} L/A$	L^{-1}	
Electric current	$I_{el} = V_{el}/R_{el}$	L	voltage V_{el} constant
Electric Power	$P_{el} = R_{el}I_{el}^2 = V_{el}^2/R_{el}$	L^{-1}	V_{el} constant
Joule effect	$W = R_{el} I_{el}^2 t$	L	V_{el} constant
Per unit area	W_{un}	L^{-1}	V_{el} constant
Electric field	E_{el}	L^{-1}	V_{el} constant
Capacity of a condensator	$C = \varepsilon_0 A/d$	L	Parallel plates
Charge of a condensator	$Q = C \cdot V_{el}$	L	V_{el} constant
Electric energy	$W_{el} = \varepsilon \cdot \varepsilon_0 E^2 V/2$	L	V_{el} constant
Stocked energy	$E_{cap} = Q^2/2C$	L	V_{el} constant
		L^3	Constant charge density
Force between plates	F_{cap}	L^2	
Magnetic field in a solenoid	$B = \mu I_{el} n/L$	L	n constant ; constant current density : $I_{el} \sim L^2$
Magnetic energy in a solenoid	$E_{magn} = B^2 \cdot V/2\mu$	L^5	
Magnetic force	F_{magn}	L^4	
Thermal energy	E_{th}	L^3	
Thermal dissipation : dissipated power	P_{diss}	L^2	conduction, radiation
Times for temperature homogeneization	τ_{th}	L^2	
Melting temperature	T_m	$1 - f/L$	f ≈ 1 nm for 3d and 5d elements

3. From micro- to nanomaterials

When one goes below the micron size, one attains the nanotechnology domain. In this range, fundamental questions appear, like : do nanomaterials behave like macromaterials ? The question is not yet fully understood. This is due to the fact that :

a) one may question the validity of thermodynamics ;
b) the number of atoms at the surface of the particles cannot be neglected as compared with the total number of atoms of the particle;
c) quantum effects appear.

Let us address these three points.

3.1. Thermodynamics

One generally considers that thermodynamics is valid when the number of atoms is « large ». What is "large" ? Let us look at the heat flow equation. It gives a macroscopic description of a material, in a volume where thermal fluctuations are « small ». But: What is "small" ? Let us consider a cube (of volume equal L^3), with N atoms per unit volume. The relative temperature fluctuation within the cube is $\delta T/T$ $(NL^3)^{-1/2}$, or L $(\delta T/T)^{-2/3} N^{1/3}$. Let us assume that the temperature is uniform when it fluctuates by less than 10^{-3}. In solids and liquids, N $10^{29} m^{-3}$, so that L 0,01 µm. In gases, N $10^{24} m^{-3}$, and L 1 µm. Therefore, the use of the heat flow equation is not valid when the size of condensed systems is less than 0.01 µm. For gases at atmospheric pressure, the critical dimension is $L < 1$ µm, i.e. it compares with the molecular flow transition, as seen before.

However, it is worth noting that there exist cases where temperature gradients are very high but the heat flow equation works. For instance, under high power laser irradiation, one calculates grad $T > > 10^7$ K·cm^{-1} , or $\delta T = 1$K over $L << 1$ nm, where the heat flow equation appears to give correct results [6,7]. It seems that the Occam'razor principle applies here.

3.1.1. The definitions of temperature

In the previous discussion, we have swept an important concept "under the rug". What is the temperature ? and: Can the temperature (an equilibrium concept) be invoked in a non equilibrium process, like heat flow ? The reader interested by this question may look to the careful analysis of the problem by Cahill et. al. [8].

The usual definition of temperature is related to the average energy of a system of particles. This definition is for a system in equilibrium and works even for nanoscale systems. *It is then valid for the study of nanoparticles in thermal equilibrium.*

In non equilibrium processes (like heat conduction), the problem is to define temperature in the presence of a temperature gradient. This is not difficult in macroscopic systems, where it is possible to define a local temperature in each space domain. The temperature might vary from one domain to the next. When small systems are considered, the main problem is to define a local temperature. There are three ways to define a local temperature.

The first definition is used, for instance, in molecular dynamics. In this method, one calculates the position and the velocity of each atom in each time period. One then calculates

the mean kinetic energy, E_c, over N time steps. N must be high enough, so that the result is statistically valid. The calculations are classical. The temperature is classically given by :

$$\langle E_c \rangle = \frac{1}{2}\langle m v_i^2 \rangle = \frac{3}{2}kT$$

The temperature is defined locally, over one atom. This approach neglects quantum effects.

The second approach takes quantum effects into account. The collective motion of atoms is described by the phonon model. They are characterised by their pulsation, $\omega(\mathbf{pol},\mathbf{q})$, depending on the wavevector, $\mathbf{q}$ and polarisation, $\mathbf{pol}$. The mean kinetic energy is given by :

$$\frac{1}{2}\langle m v_i^2 \rangle = \sum \left\{ \hbar\omega(\mathbf{q}) / \left[\exp\left(\hbar\omega(\mathbf{q})/kT - 1\right) \right] \right\}$$

At high T (well above the Debye temperature), the classical and quantum equations give similar results. However, at low temperature (and near room temperature), both definitions give different values of T. A third definition is to substract the zero temperature motion from the quantum definition.

Among the three definitions, which one is the correct one? It depends on the size of the domain where T is defined. The classical definition is purely local. T may be defined for each atom or row of atoms.

With quantum definitions, the length scale is defined by the phonon mean free path. If two space domains are characterized by different temperatures, the phonon distributions are different. One domain is defined by one phonon distribution. Hence the characteristic length of a local domain at a given fixed T must be larger than the phonon diffusion length. But this length is a function of the phonon frequency. Low frequency phonons have large mean free path, while high frequency ones have shorter mean free paths. From this point of view, T may not be defined on one atom or row of atoms. In particular, one may not have abrupt variations of T between two successive rows of atoms.

However, molecular dynamics calculations of heat flux through grain boundaries show that one might observe sharp temperature variations through grain boundaries. As discussed by Cahill et. al. [8], this might mean that grain boundaries are a natural border between domains where T are different.

3.1.2. Heat transfer

In ordinary matter, conduction heat transfer is described by the Fourier's law, stating that heat flux is proportional to the temperature gradient between the extremities of the material and to the thermal conductance. This is not valid for nano-objects. Indeed, the mean free path of phonons is generally much larger than the size of nano-objects. Then, phonons behave in a ballistic way, without collisions within the nano-object. So the heat transfer is more rapid than under usual conditions. A consequence is that the time of heating of nanosystems is shorter than predicted by classical heat transport equations. For instance, a cube of dimension 10 nm attains equilibrium in about 10^{-12} s.

Particles with a diameter in the range of 1 - 100 nm are in an intermediate state between the solid and the molecular ones. Such particles are characterised by the fact that the ratio of the number of surface to volume atoms, A, is not small. For instance, for a particle containing about 4000 atoms, of radius, $R \approx 2$ nm, one calculates that $A \approx 0.3$. It is then obvious that the effects of the surface on the cohesive properties of the particle cannot be neglected. This is seen in various situations, like the size-dependent melting point depression [9,10] and other phase transitions [11,12] of nanoparticles. Since the surface tension depends on the chemical environment, it is obvious that the melting point and phase diagrams variations depend on it, as observed experimentally for various cases [13, 14, 15].

In inorganic materials, the melting temperature, T_m, varies with the radius of the particle, R, like ($T_{m\,\infty}$ is the bulk melting point) :

$$T_m = T_{m,\,\infty} [1 - \alpha/(2R)].$$

α depends on the material. It is between 0.4 and 3.3 nm [10]. A few examples are given in Table 2.

Table 2. Coefficient of variation of T_m for a few elements

Element	$T_{m,\,\infty}$ (K)	α (nm) [10, 16]	α_{exp} (nm)
Ag	1234	1.27, 0.96	
Al	933	1.14, 1.487	0.6
Au	1336	0.92, 1.1281	0.96
Co	1768	1.00, 0.9	
Ge	1210.6	2.30,	
		3.33, 1.06	
In	429.4	1.95, 2.65	0.974
Pb	600.6	0.98,	1.048
		1.40, 1.79	
Si	1683	1.88, 0.84	
Sn	505.1	1.57, 2.278	1.476

There is also an effect of size on the phase diagrams of nanoparticles. Again, this is due to the effect of the surface on the cohesion of the particle [13].

Since the number of surface atoms play a role, it is obvious that the phase diagram also depends on the shape of the particle. It is demonstrated [17] that the phase diagram of non spherical particles may be calculated from the spherical case, at the corresponding value of the ratio surface area vs. volume, both without and with surface segregation, provided the surface tension is isotropic. This is important when evaluating the effect of the shape on the materials properties.

When looking at the phase diagram of nanoparticles, one has also to take into account the fact that the total number of atoms within one particle is limited. This is demonstrated to lead to various particular effects. In particular, when one looks at the nucleation of one new "crystal" phase of the nano-particle, it turns out that there exist three possibilities: phase separation, prohibition of decomposition and formation of metastable state of nanoalloy [18].

This reasoning is valid in the thermodynamical limit, i.e. when the radius is of the order of 3 nm or above. When the number of atoms, N, in the nanoparticle decreases down to 100 or below, the stability is calculated by molecular dynamics methods [19]. It turns out that the melting and solidification temperatures are different. At low temperature, the particle is "solid": the positions of the atoms don't vibrate much. At high temperature, the particle is "liquid": the mean values of the interatomic distances fluctuate much around the equilibrium positions. At intermediate temperature, things depend on the directions of the temperature change. When one starts from low temperature and the system is heated, melting takes place at a given temperature. When one cools down the particle from the liquid state, the solidification occurs at a solidification temperature, different from the melting one. It is worth noting that this also means that it is possible to define the "solid" and "liquid "states for very small nanoparticles.

3.2.1. Matter in nanotubes

The flow of liquids in nanotubes is now an interesting field of research. In particular, so-called carbon nanotubes are seen as potential submicroscopic test tubes or pipes. Their interest is tremendous to obtain nanothermometers (by using the dilatation of bubbles in nanotubes), or hydrogen containers in future hydrogen reservoirs in cars equipped with fuel cells. For such goals, it is necessary to understand how nanotubes and liquids interact, in particular the wetting properties. In classical models, this is described by the Young-Laplace equation:

$$\Delta p = (2\gamma/r) \cos\theta,$$

where Δp is the pressure difference across the liquid-vapor interface in a capillary, r is the radius of curvature of the meniscus, θ is the contact angle between the meniscus and the surface. The liquid is drawn spontaneously into the capillary, when Δp is positive. This implies that $\theta < \pi/2$. It is hard to predict the value of θ. Experimental results indicate that carbon nanotubes are wet only by low-surface-tension liquids [19]. This is in qualitative agreement with the macroscopic behaviour. When the liquid does not wet the nanotube, other methods have to be applied, like their opening by wetting first by acids.

Table 3. Wetting properties of carbon nanotubes [20]

Substance	Surface tension (mN/m)	Wetting
HNO$_3$	43	Yes
S	61	Yes
Cs	67	Yes
Rb	77	Yes
V$_2$O$_5$	80	Yes
Se	97	Yes
Te	190	No
Pb	470	No
Hg	490	No
Ga	710	No

It seems intuitively obvious that only liquids might wet carbon nanotubes. However, it has been noted above that the melting temperature, T_m, is both a function of size and shape. It may be expressed following [17] :

$$T_m = T_{m,\,\infty}\,[1 - (\alpha/6)(A/V)].$$

(A/V) is the ratio between the number of surface and volume atoms. It is calculated by purely geometrical arguments. It is worth noting that, since the sphere is the geometrical figure with the lowest (A/V) at constant V, the size variation of all other 3-dimensional shapes is larger than for the sphere. One may express :

$$A/V = \beta(\text{shape}) / V^{1/3}$$

Table 4. Parameter β for various geometrical shapes

Shape	β(shape)
Sphere	4.836
Cube	6
Octahedron	5.719
Icosahedron	5.149

In nanotubes, the inner matter is cylindrical. In this case, it is easy to demonstrate [21] that the ratio α(sphere) : α(cylinder) is equal to 3 : 2.

Whatever this, it turns out that material within the nanotube might melt at lower (or higher) temperature than the bulk melting point. For instance, it has been shown experimentally that the crystallinity of a Pb wire of diameter 3 nm in a carbon nanotube disappears at room temperature [22]. This is in semi-quantitative agreement with the theoretical models. They predict that, under these conditions, T_m is in the 360-450 K range.

3.3. Quantum effects

Quantum effects manifest themselves mainly when one studies the electronic and optical properties of nanosystems. The size of the systems where quantum effects are important depends on two parameters [23,24] :
-   the de Broglie wavelength of electrons : λ_{el} ;
-   the Bohr radius of the exciton (electron-hole pair), a_B, in semiconductors:

$$a_B = \varepsilon \hbar^2 / \mu e^2$$

ε is the dielectric constant, μ is the reduced mass and e is the charge of the electron. These dimensions are of the order of a few nm. At such small dimensions, the optical and transport properties of nanoparticles are different from the ones of the bulk materials. For instance, the optical absorption edge of semiconductors is shifted "to the blue" when L decreases.

One also may design "mesoscopic" structures, for which one, two or three dimensions compares or are smaller than λ_{el} and a_B. In these structures, the elementary excitations are

subject to a quantum confinement. When the confinement involves one direction, while the two others are infinite, the structure is said to be 2-dimensional. It gives rise to a so-called quantum well. When confinment is in two directions, one obtains a quantum wire. When there is a quantum confinement in three directions, one has a quantum dot. The electrical and transport properties of these systems differ from the ones of the bulk material.

As said previously, when one considers transport in nanoscale systems, the question of the nature of transport at the atom size has to be carefully addressed. At the boundary between two "grains", the temperature is not defined. So, how do electrons or heat jump from one side to the other ? In these cases, one often adopt the so-called Landauer formalism. Within this formalism, one assumes that there are two reservoirs (of electrons or heat) at known temperatures. The flows are considered to be ballistic. One then computes the energy flows through the possible channels between the two reservoirs. It turns out that the electron and heat flows are quantized.

The electron flow is described by the electron conductance, G, given by the so-called Landauer formula :

$$G = (e^2/\pi\ )T/(1 - T).$$

T is the transmission by channel [25]. Experimentally, one observes that the conductance increases by integer values of the quantum of conductance ($e^2/\pi\ $). Using similar theoretical arguments with phonons, at low temperature, it has been shown that in a ballistic, one-dimensional channel, there exists a quantum of thermal conductance (T = temperature) [26]:

$$G_{th} = \pi^2 k_B^2 T/(3h)$$

4. Conclusions

Altogether, it turns out that our intuition and knowledge of the behaviour of materials at the macroscopic scale is not well adapted to the understanding of nanomaterials. The use of scaling laws is useful, but not sufficient. When going to the nanoscales, questions relevant to the validity of thermodynamics, size effects on the cohesion of particles, quantum effects have to be carefully addressed. It turns out that these three questions begin to manifest themselves when the characteristic dimensions of the particles decreases down to the 5-10 nm range. The combined effects of cohesion and quantum effects has not yet been addressed to our knowledge.

Acknowledgments

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