Physics of Plasmas

Preface to Special Topic: Plasmonics and solid state plasmas

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Preface to Special Topic: Plasmonics and solid state plasmas

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Plasmonics, the study of the interaction of electromagnetic radiation with electrons in solids, is an exciting new field that has developed fast since the 1980s and is still growing steadily. Yet, plasma physicists have devoted little attention to it. This special collection would like to bridge the gap between plasmas and plasmonics and encourage plasma physicists to have their say in this burgeoning research field. *Published by AIP Publishing.* https://doi.org/10.1063/1.5026653

Plasma physics as a distinct scientific discipline is less than one century old. Indeed, Irving Langmuir coined the term *plasma* in his seminal paper published in 1928[1] and defined a plasma as a "region containing balanced charges of ions and electrons." Since its beginning, the development of plasma physics has been driven principally by potential applications to three different fields: (i) Thermonuclear fusion for both military and peaceful purposes, (ii) astrophysics and space sciences, and (iii) engineering and industrial applications of low-pressure plasmas. To this date, these three fields account for most publications in the area of plasma physics.

If we stick to Langmuir's original definition, some form of plasma should be ubiquitous,[11] because ordinary matter is neutral, obviously containing (roughly) "balanced charges of ions and electrons." Arguably, a further reasonable requirement would be that at least some of the charge carriers are mobile, in other words that the plasma medium possesses some dynamical properties. This is the case for conduction electrons in metals, which behave to some extent as free electrons moving in the presence of a neutralizing ion lattice. It is a rather crude approximation, but it tells us that some plasma-like properties are to be expected when studying the dynamics of electrons in metals.

Perhaps the most salient feature of ordinary plasmas is that they display collective effects, i.e., effects that depend not only on local conditions but also—indeed principally—on the properties of all the particles in the system. Such collective behavior is possible because of the long-range nature of electromagnetic forces, and hence, it should be expected for electrons in metals too. In 1956, Pines[2] coined the term *plasmon* to denote the collective oscillations of conduction electrons in a metal, stressing the similarity with other collective oscillations occurring in gaseous plasmas. *Plasmonics* may be defined as the study of the interaction between electromagnetic radiation and free electrons in a metal and all the accompanying collective phenomena.[3] As a separate topic, plasmonics evolved very rapidly since its early debuts in the 1980s and 1990s. However, it developed in parallel, and without any significant interactions, with traditional plasma physics. It is fair to say that the vast majority of plasma physicists have largely ignored this burgeoning field of research. This is a pity. One of the purposes (or hopes) of the present Special Topic issue is thus to foster fruitful scientific exchanges between the plasmonics and plasma physics communities.

Two important differences between ordinary gaseous plasmas and solid state plasmas should nevertheless be mentioned.[4] First, plasmonic effects rely strongly on the presence of interfaces (metal-vacuum and metal-dielectric). Plasmon modes are often surface modes, either localized at the interface or propagating along the surface. For this reason, plasmonics is to some extent a branch of nanophysics, and most relevant objects studied and utilized for plasmonic applications are nanoscale objects, such as nanoparticles or thin films. The word *nanoplasmonics* has been coined to stress this fact.

Second, the electron density in metals is very high, of the order of $n_e \approx 10^{28}\,\mathrm{m}^{-3}$. At those densities, the electron gas is fully degenerate even at room temperature, so that it is governed by the Fermi-Dirac statistics and displays quantum properties; quantum effects are really inescapable for solid state plasmas, although some level of semiclassical modelling is still useful and used. Another consequence is that the corresponding plasma period is very short, $2\pi/\omega_{pe} \approx 1$ fs. It is no surprise then that the development of plasmonics coincided with recent advances in ultrafast optics, such as the availability of femto- or attosecond laser pulses. Finally, high density also means that the electron gas is strongly correlated, so that mean-field approximations (the analog of the Vlasov-Maxwell equations for gaseous plasmas) are not entirely appropriate.

In summary, if one had to capture the essence of solid state plasmas and plasmonics in just four words, it would be fair to choose nanometer, femtosecond, quantum, correlations.

As an illustrative example, let us consider a localized surface plasmon mode in a spherical metallic nanoparticle (Fig. 1). The mode is excited by an ultrafast laser pulse in the visible range (400–800 nm), so that the electromagnetic wave length is much larger than the diameter of the nanoparticle. The electric field of the laser drives the electrons away from their original steady state. The Coulomb force exerted by the ion lattice tends to bring the electrons back to equilibrium, but due to their inertia, they overshoot it and begin to collectively oscillate at the so-called Mie frequency

$$\omega_{\mathrm{Mie}} = \frac{\omega_{pe}}{\sqrt{2\epsilon_m + \epsilon_b}}, \tag{1}$$

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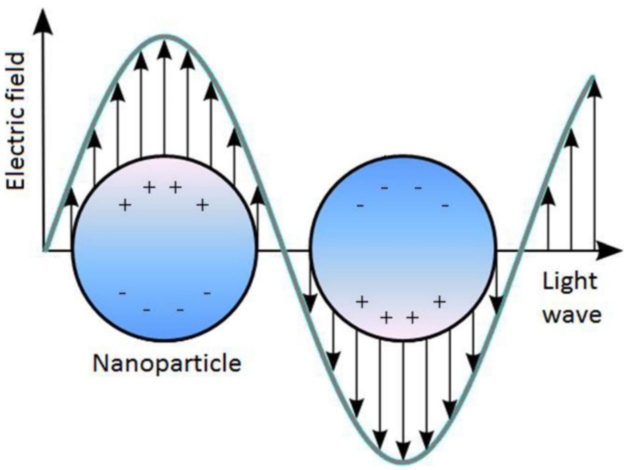

FIG. 1. Schematic representation of the localized surface plasmon resonance for a spherical metallic nanoparticle. Reproduced with permission from Hammond *et al*., Biosensors **4**, 172 (2014). Copyright 2014 MDPI.[9]

where ϵ_m is the dielectric constant of the environment embedding the nanoparticle, and ϵ_b is the dielectric constant of the bound electrons inside the particle. Taking $\epsilon_b = \epsilon_m = 1$, we obtain $\omega_{\text{Mie}} = \omega_{pe}/\sqrt{3}$, which is the standard Mie expression for plasmonic oscillations in a nanoparticle. In practice, the surface plasmon mode is observed in optical experiments on metallic nano-objects as a sharp resonance near the Mie frequency. It is called surface plasmon because the space charge is localized near the particle's surface, while the bulk remains neutral.

Of course, Eq. (1) has been obtained thanks to some very drastic approximations. We have neglected (in no particular order) thermal corrections, quantum effects, nonlinear effects, correlations, all sorts of damping (e.g., electron-electron and electron-phonon couplings and radiative damping), all effects due to the electron spin, and relativistic effects (e.g., the spin-orbit coupling). Thus, plasmonics can be viewed as the theoretical and experimental study of collective motions such as the surface plasmon mode, incorporating many or most of the above-mentioned effects, in a great variety of different geometries (see Fig. 2). The latter include non-spherical nanoparticles (ellipsoids, cubes, pyramids, star-shaped,…), hollow particles, thin films, rods, disks, and all combinations thereof, such as dimers, trimers, chains and arrays of nanoparticles, or even exotic systems like silver nanocubes lying on a nanoscale dielectric layer deposited on a gold film.[5]

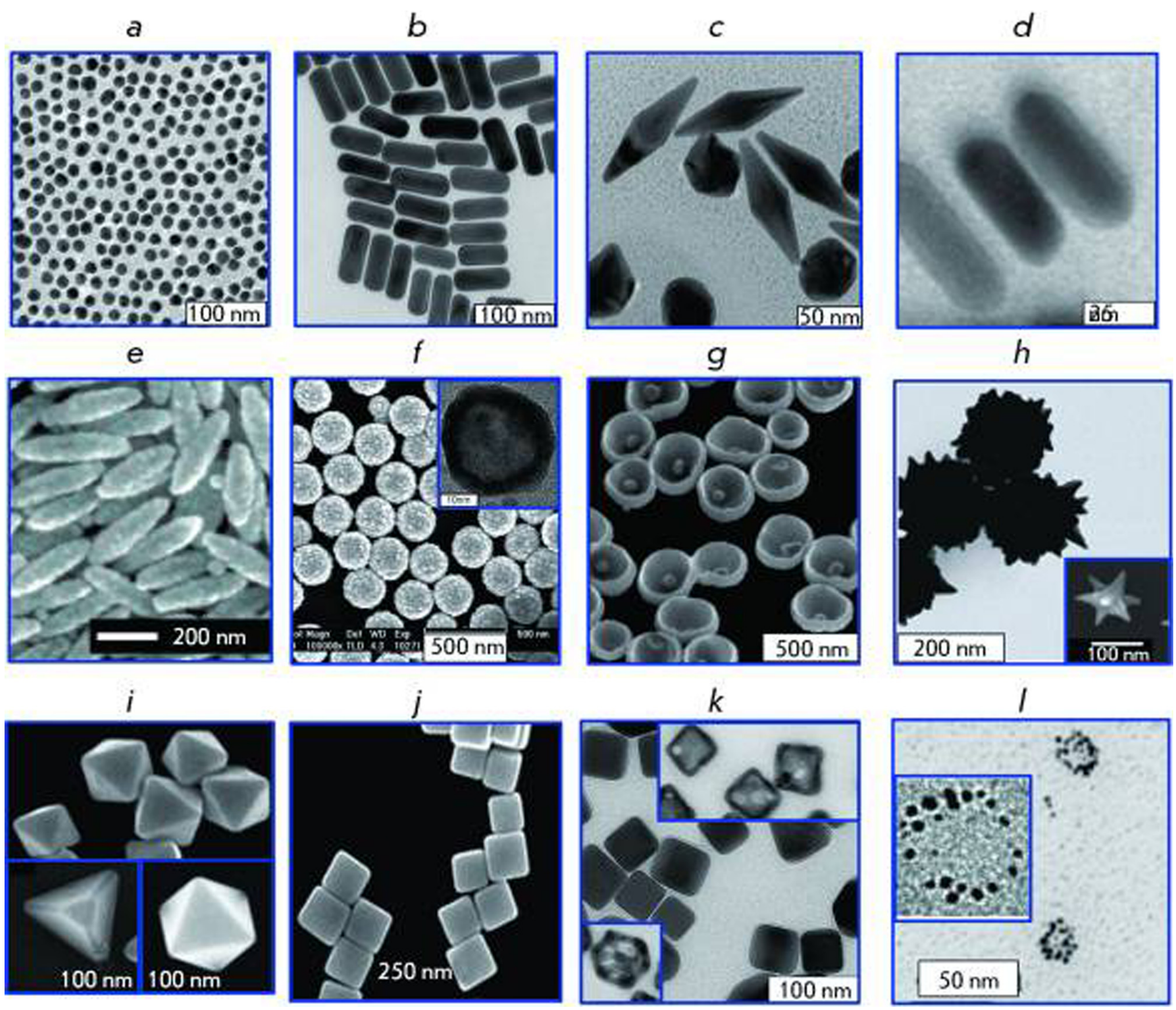

FIG. 2. Various types of gold nanoparticles. Reproduced with permission from Dykman and Khlebtsov, Acta Nat. **3**, 34 (2011). Copyright 2011 Park-media Ltd.[10]

The present Special Topic issue is a first attempt to foster cross-fertilization between the two communities of plasma physicists and experts in plasmonics. The papers published in this issue are proof that some active contacts already exist. Indeed, some of the authors are plasma physicists who ventured to explore plasmas whose properties make them relevant for plasmonics.

The most likely subarea of plasma physics where plasmonic effects are bound to play a role is laser-plasma interactions. In current experiments, plasmas are produced by ablating a solid target by means of intense laser beams. The resulting plasma displays, at least in the initial stages if its evolution, the same high density of the solid target, so that phenomena typical of solid state plasmas can be expected—although mitigated by the fact that the temperature is much higher. Here, Andrea Macchi reviews recent studies on the interaction of intense laser pulses with solid targets and their impact on the generation of propagating surface plasmons. In a closely related paper, Giada Cantono *et al*. report on the excitation of surface plasmons with ultraintense laser pulses and the concomitant production of highly collimated bunches of relativistic electrons.

Surface plasmon modes are the subject of three further contributions. The paper by Wang and Cappelli describes propagation, in a plasma slab, of novel plasmonic modes with frequency smaller than the plasma frequency. Osamura Sakai and coworkers investigate wave propagation in media with a negative dielectric constant and notice that the observed modes display some of the features of localized surface plasmons and surface plasmon polaritons on a metal surface. Smolyakov and Sternberg discuss the role of surface plasmon resonances in the reflection and absorption properties of a dense plasma slab.

Finally, two contributions are devoted to the microscopic modelling of solid state plasmas.

Most theoretical models in condensed matter physics are *wavefunction based*, like the Hartree and Hartree-Fock equations (the quantum equivalent of the Vlasov-Poisson equations). Density functional theory (DFT)[6] in its most common form (Kohn-Sham equations) is also wavefunction-based and may be viewed as an upgrade of the Hartree equations that can in principle accommodate all types of electron-electron correlations. However, more recently, alternative methods such as quantum hydrodynamics (QHD)[7] have become increasingly popular and start being utilized in the plasmonics community.[8] The work by Moldabekov, Bonitz, and Ramazanov is a timely contribution that analyzes the theoretical foundations of QHD and discusses the validity of the underlying approximations.

Wavefunction-based methods suffer from an important limitation insomuch as, being Hamiltonian, they cannot easily incorporate dissipative effects. In their contribution, Vincendon, Reinhard, and Suraud try to lift this limitation and address the impact of damping and dissipation on plasmonic modes using time-dependent DFT augmented by a quantum relaxation time approach.

After over three decades of fast development, plasmonics is by now a mature discipline. But, although many aspects of it are well understood, there is still room for contributing to its advances with the experience accumulated in the plasma physics community. Let me just mention one possible avenue for new research. Current plasmonics focusses on either very small objects, for which quantum wavelike effects are important, or very large nanoparticles (diameter > 100 nm) that contain a great many electrons. In the latter case, microscopic approaches such as DFT or Hartree-Fock rapidly become too heavy in terms of computational cost. One alternative is to turn to macroscopic models, such as the QHD approach mentioned above. But plasma physicists have developed over the years a whole battery of intermediate reduced models—e.g., guiding center, gyrokinetics. Some of these methods might be transposed with profit to the field of plasmonics, perhaps with some significant modifications.

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[11]We used to tell our students that plasmas represent 99% of the universe content. Since the advent of dark matter (27%) and dark energy (68%), plasmas have dwindled to a mere 5%. Not a good omen.