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## Plasmonic semiconductor nanoparticles showing nonlocal response

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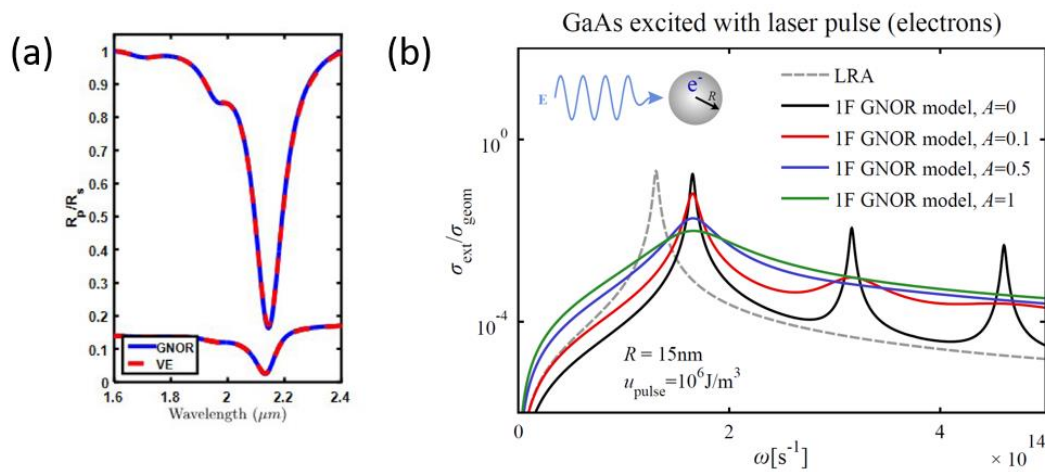
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**Abstract.** We predict that localized surface plasmons (LSP) in semiconductor particles exhibit spatial nonlocal response effects as the geometry enters the nanometer scale. To investigate these nonlocal effects, we first apply the hydrodynamic model (HDM) to nanospheres of two different semiconductor materials: intrinsic InSb and n-doped GaAs. Our results show that the semiconductors indeed display nonlocal effects, and that these effects are even more pronounced than in metals, and more tunable as well. We also present a two-fluid hydrodynamic model for semiconductors containing electrons and holes (from thermal or external excitation) or light and heavy holes (in p-doped materials). The two-fluid model predicts the existence of two longitudinal modes, an acoustic and an optical, whereas only an optical mode is present in the HDM. By extending nonlocal Mie theory to two plasmas, we simulate the optical properties of two-fluid nanospheres and predict that the acoustic mode gives rise to peaks in the extinction spectra that are absent in the HDM. And from a numerical study, we predict that by considering dimers rather than monomers of nanowires, the extinction cross section and field enhancement of the acoustic localized surface plasmon resonances can increase substantially. In this conference proceedings, we present calculations of the two-fluid GNOR model, which show that acoustic surface plasmon modes are surprisingly robust against size-dependent broadening.

The interest in the research field of nanoplasmonics arises because plasmon wavelengths can be much smaller than optical wavelengths, and because electric fields can be hugely enhanced near metal structures. This enables miniaturization of optical components and strong light-matter interactions, of metal structures interacting with quantum emitters in particular [1,2]. Electronic circuits have become much smaller in past decades and the interest to also make optical circuits and devices smaller is fueled by ambition to replace electronic by photonic circuitry in information processing. The question arises how small one could make plasmonic particles and circuits. Currently plasmonic particles of sizes of the order of one nanometer and gaps well below a nanometer can be fabricated, and here the usual description of the plasmonic material in terms of a refractive index breaks down. Instead of using classical electrodynamics, one should at these subwavelength scales take new physical mechanisms into account such as nonlocal response and electronic spill-out. Nonlocal response is the phenomenon where the electric polarization at a certain position in a nanoparticles depends on the electric field also in its vicinity; in infinite media nonlocal response can be described by a response function that depends not only on frequency (dispersion) but also on wave vector (spatial dispersion) [3-5]. The other phenomenon beyond classical electromagnetism, electronic spill-out, is a consequence of the electrons not being perfectly contained within the geometric boundaries of a metal structure, whereby the free-electron densities have decaying tails on sub-nanometer scales across the metal boundaries. Nonlocal response is most often described within a semiclassical hydrodynamic Drude model, which contrary to the simple Drude model (“local-response approximation” or LRA) takes longitudinal pressure waves within the free-electron plasma into account [5]. Spill-out can be taken into account as well using so-called self-consistent hydrodynamic models [6], but this will be neglected in the following.



Nonlocal response of metal nanostructures is an active research topic, both experimentally and theoretically. One signature of nonlocal response in noble metal structures is a spectral blueshift as the size is decreased. It has long been known [7] that there is also a concomitant spectral broadening, phenomenologically described for nanospheres as a size-dependent damping constant  $\gamma' = \gamma + A v_F/R$ , where  $\gamma$  is the Drude damping constant,  $v_F$  the Fermi velocity,  $R$  the radius of the nanosphere, and  $A$  is a dimensionless constant that for noble metals turns out to be of order unity. Recently this spectral broadening was also identified as an effect of nonlocal response: generalized nonlocal optical response (GNOR) theory described the broadening as well as the blueshift [8], and  $A$  turns out proportional to the diffusion constant of GNOR theory. As a third effect of nonlocal response, new resonances may be found above the plasma frequency, which can be interpreted as standing waves of bulk plasmons, longitudinal resonances of the hydrodynamic pressure waves.



**Figure 1:** (a) Prediction of visco-electric theory of Ref. [10] (well reproducing reflectivity measurements in Fig. 1 of Ref. [10]), here overlapping with our GNOR prediction [12]. Small oscillations on the short-wavelength shoulder correspond to standing bulk plasmons. (b) Extinction cross section for a semiconductor nanosphere calculated in the usual single-fluid (1F) GNOR model, for various levels of size-dependent broadening. The 1F GNOR model with  $A=0$  corresponds to the HDM. Adapted from Ref. [11].

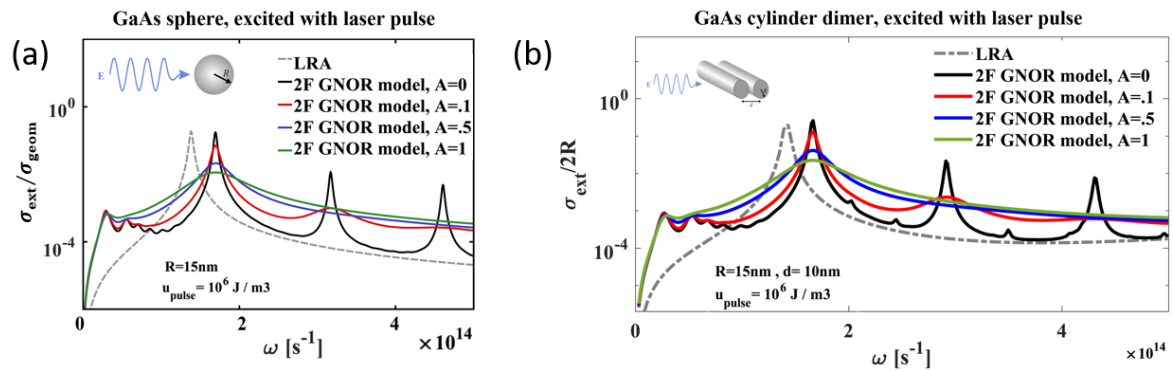
Recently some of us proposed that effects of nonlocal response may not only be observable in metals, but also in semiconductor structures that are either optically or thermally excited [9]. Semiconductors are certainly “smarter” plasmonic materials than metals, since their free-electron densities are tunable, which enabled an amazing semiconductor industry. We proposed that nonlocal-response effects in semiconductors would occur in intermediate-size structures, too large for a description of single-particle quantum confinement (as for quantum wells, wires, or dots), but too small for the usual bulk description to be valid either. Our calculations showed that relative plasmonic blueshifts will typically be much larger in semiconductors than for metals [9]. Very interestingly, since then such hydrodynamic effects have indeed been observed, in planar semiconductor structures made of indium-doped cadmium oxide (In:CdO) [10]. In particular, both the mentioned nonlocal blueshift and the broadening of spectra for smaller structures were seen, as well as standing bulk plasmons. It is an interesting open question in what further semiconductor structures these phenomena can be observed. We have checked that the size-dependent broadening as observed and explained by a visco-electric (VE)

theory in [10] is equally well described by the GNOR theory, if we allow the GNOR diffusion constant to be a fitting parameter  $D = 10^{-4} \text{ m}^2/\text{s}$  (corresponding to  $A = 0.1$  for spheres), see Fig. 1(a). This diffusion constant for a layered semiconductor is roughly an order of magnitude smaller than in a noble metal nanosphere [11,12]. Extinction spectra for semiconductor nanospheres with various amounts of size-dependent broadening are presented in Fig. 1(b).

There may be more subtle hydrodynamic effects in semiconductors that have no counterpart in metals. For example, the existence of more than one free-carrier plasma may give rise to new observable nonlocal effects [13], see also Refs. [14–16]. Here one can think of systems with an electron plasma plus a hole plasma, or p-doped systems with both heavy-hole and light-hole plasmas. We proposed to describe this by a two-fluid hydrodynamic model. One might expect that such a model would simply predict the combined effect of surface-plasmon polaritons (SPPs) of the one plasma with SPPs of the other plasma, but the situation is more interesting. Recall that the usual surface-plasmon polaritons are hybrid excitations of both light and the free-electron plasma, with energy swapping back and forth between light and matter. In the two-fluid model however, the light “sees” two oscillating plasmas, and the simplified story is that these plasmas can oscillate with their current densities either (almost) in phase or (almost) out of phase. This leads to new plasmon resonances besides optical ones: acoustic plasmons.

Emergent new behavior is that the plasmons corresponding to almost-out-of-phase current-density oscillations occur at frequencies lower than either of the plasma frequencies of the individual fluids. So low-energy peaks occur where the usual single-fluid theory predicts none. These plasmons are the acoustic plasmons, and in calculated spectra they are therefore the most characteristic signature of the two-fluid model. The plasmons corresponding to in-phase oscillations of the current densities of the individual plasmas are optical plasmons. If we consider the special case of two identical plasmas with the same damping constants and nonlocal parameters, then our two-fluid model reduces to the usual single-fluid hydrodynamic model with only the optical plasmons [13]. However, in the general two-fluid model there are many more resonances in the spectra, and for simple geometries they can still all be classified. We have studied the optical and acoustic surface and bulk plasmons both for semiconductor nanospheres [13] and cylinders [16], and we found similar results for both geometries. Moreover, we have numerically implemented the two-fluid model in COMSOL Multiphysics, a commercial FEM package, and benchmarked it against analytical calculations for cylinders [16]. This allowed us to study the characteristic acoustic resonances in more advanced structures such as dimers. Gap structures are well-known for their capability to amplify electric fields, and indeed we found that the acoustic resonances are enhanced for dimer structures, although the gap enhancement is not as spectacular as for the usual optical surface plasmons.

One thing that we did not yet take into account in our previous publications on the two-fluid model [13,16] is size-dependent broadening. The question is whether the resonances in the two-fluid model will still be visible when size-dependent broadening causes the resonances to broaden and flatten. It is especially interesting whether the characteristic acoustic resonances will still be discernible when going from a two-fluid HDM to a two-fluid GNOR model. In Figure 2(a) we show extinction cross sections for an optically excited GaAs nanosphere, with parameters as in Ref. [13], but now for four values of the  $A$  parameter, where the two-fluid GNOR model with  $A=0$  reduces to the two-fluid hydrodynamic model of Ref. [13].



**Figure 2:** (a) Extinction spectrum of a semiconductor nanosphere as in Fig. 1(b), but now modelled using the two-fluid (2F) model, taking both the electron and hole plasmas into account. (b) Extinction spectrum for a dimer of cylindrical nanowires, as in Ref. [16], but now within the two-fluid GNOR model, for various strengths of the size-dependent broadening. In both panels, the lowest-energy resonance is the acoustic surface plasmon resonance. The local response approximation (LRA) corresponds to the Drude model.

As is clear from Figure 2(a), size-dependent broadening affects all resonances to some extent, but the lowest-energy peak, which is the acoustic surface plasmon peak, is rather robust, in that it does not disappear as the value of  $A$  is increased. Fig. 2(b) shows that the same holds for a dimer of nanocylinders: also here, several pronounced peaks disappear almost completely in the two-fluid GNOR model, but the acoustic peak remains rather robust against size-dependent broadening. On the other hand it must be said that the vertical axes are logarithmic, and it will be more challenging to measure the acoustic SPP than the optical SPP (highest peaks).

In conclusion, we suggested that plasmonic properties of some mesoscopic semiconductor structures might be described by the same hydrodynamic Drude model as used for metals, as has been experimentally confirmed. We also developed a two-fluid hydrodynamic Drude model suitable for semiconductors with two types of plasmas. In this model, new acoustic resonances appear that have not yet been observed experimentally, and which are enhanced in dimer structures. We here presented calculations of the two-fluid GNOR model, which show that acoustic surface plasmon resonances are surprisingly robust against size-dependent broadening.

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