Perspective on plasmonics

Joachim Krenn was one of the early pioneers of modern surface plasmon optics, and has almost 8,000 citations to his work. *Nature Photonics* spoke to Krenn about the field's origins.

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When did plasmonics start and how did we arrive at modern plasmonics?
Important contributions to what later became called plasmonics can be traced back to quite a while ago. Gustav Mie with his theory in 1908 is one example. The Maxwell–Garnett work a couple of years before that is also relevant. These works gave many descriptions of the nanoparticle-related effects that we know today. There have not been that many changes to the theory since then; it required just a few extensions. At around the same time or even earlier, there was some beautiful experimental work, which is largely overlooked, by the Austro-Hungarian scientist Richard Adolf Zsigmondy. He invented the slit ultramicrocope — a dark-field microscope — and noticed the colour of gold particles in a colloidal solution. He knew he was looking at the scattering from individual gold nanoparticles.

In 1925, Zsigmondy was awarded the Nobel Prize in Chemistry for his work on colloids. In the 1950s, there were relevant works related to electron energy loss in metals by Ritchie and Powell, as well as Pines and D. Bohm, which were followed by the well-known works of Kretschmann, Raether and Otto in the 1960s.

From there we could consider the historical perspective from the point of view of the activities, at the University of Graz, led by Franz Aussenegg, from whom I 'inherited' the field. In the 1970s, he was doing molecular spectroscopy and surface-enhanced Raman spectroscopy was published by several groups. He looked into this new direction but very quickly realized it was complicated, with many ingredients and a lack of control over them. However, he was fascinated by the nanoparticles and left the molecules. This led to nonlinear work on the nanoparticles and biosensing research, and then in the 1990s scanning tunnelling microscopy was used as well. In the mid 1990s, he decided that these colloids and 'island' films were still too complicated. He wanted more control over the geometry.

This was when I was a student here, and at the same time Aussenegg decided to get an electron-beam lithography set-up. It was a long route to move from the complicated system to the single particle, which Zsigmondy was already seeing over 100 years ago. This path probably occurred for a lot of people working during the early stage of the modern era of plasmonics. Many people came from interesting but complicated systems and then arrived in the 1990s with all of these new instruments (near-field scanning optical microscopy, femtosecond-resolved spectroscopy and so on), to do work on individual nanoparticles and controlled structures. But also, they were ready to do it.

Why did so many researchers come to the field in the last decade?
I cannot say why you and other people came into plasmonics in the early 2000s. However, I think almost everything you find in photonics can be considered in terms of plasmonics, and so potential applications are everywhere. So far, plasmonics has not really lived up to our dreams that we had back then, but there are still several good reasons for our interest.

Plasmonics can provide nanoscale confinement and localization. This is related to waveguiding, coupling to emitters in excitation and emission, and active functionality on the nanoscale, and you can find applications to just about every aspect of photonics. People also started to realize that apart from spectroscopy and nonlinear effects, plasmonics might also have other uses. In particular, there are works from the 1970s and 1980s discussing plasmon-guided light on thin films and cylinders. Although subdiffraction-limited localization of guided modes by some structures is certainly implied in many of the early works, one of the first explicit proponents of this application was Junichi Takahara, in 1997.

Regarding the high spatial localization and the density of states, I do not really know of other systems that can do it so well on the same scale. The features allow us to change the emission and excitation properties of emitters, and I think there is a long way to go before we really exploit that to its full power; but radiative rate engineering is already well established. These small mode volumes and high Purcell factors are really big reasons for using plasmons.

Tell us about some of the main challenges.

There are also some challenges and problems too. We cannot control the surface geometry very well. If you look closely, you have grain boundaries, roughness and other imperfections. And most of us are not always characterizing this and instead assume a smooth surface. There is no good definition of metal in other words. Also, if we want to use some emitter near this poorly defined surface, then its distance or orientation with respect to the surface is not well known, on the single-digit nanometre scale.

The same goes for the surface chemistry. If we want to exploit these highly localized electromagnetic fields fully, especially when it comes to gaps or coupled nanostructures, we have to learn how better to control the materials and the overall experimental conditions.

A few nice approaches to this problem exist, already dating back several years; for example the use of scanning probes with single particles attached to them (see, for example, the work of Novotny and Sandoghdar) and scanning single molecules. However, such an approach would have much more potential and impact if we could learn how to attach not only a simple sphere to the probe, but also other plasmonic structures with tailored resonances.

Also, very good control over monocrystalline surfaces would allow us better to get down to the scale where quantum effects are expected to become significant, as indicated in recent works by Jen Dionne and others. However, to include these quantum aspects into our research we need spatial control over all ingredients that is maybe one order of magnitude better than now.

What about the ongoing debate regarding large losses, and compensating gain?

We are somewhere in the middle of the story of plasmons and loss and using gain to counteract it. The first efforts involved dye molecules in a surface plasmon resonance set-up, but only weak effects were observed. The systems have evolved, and in recent years the subfield has changed a lot, up to the claim of lasing. Some of the results are quite convincing, showing that stimulated emission can contribute some gain to the system, but I think there is still some controversy about actual lasing activity.

One problem is that the definition of plasmon lasers, or spasers (and even lasing spasers), has blurred somehow. Another point is that some of the systems are scaled versions of what has already been done elsewhere, for example in quantum cascade lasers at 3–5-μm wavelength or in conventional semiconductor lasers at visible wavelengths.

The question also arises of whether simply having metal surfaces or cladding qualifies something to be a plasmon laser. Do you need to pump a surface plasmon that then emits? And so on. As I see at conferences, some confusion exists. Of course, we would really like to see the surface plasmon being pumped, and this surface plasmon would be subdiffraction-limit localized to differ from what can easily be done with conventional lasers. Also, clear indication of lasing from the threshold characteristics, spectrum and output is essential.

The problem is that as your surface mode becomes more localized, the loss increases and, hence, the gain requirements can be high for some structures. For many subdiffraction-limit sized modes, there is not a medium available that can provide sufficient gain. However, quantum dots seem to be one of the best options for this application. There are also options for improving pumping schemes.

Are people being realistic about the claims they make and the applications?

When you are in a field for a long time, sometimes you seriously doubt if the works are really that novel and wonder whether there is hype. However, I think there are still new things going on and surprises around the corner. Originally, who would have predicted the possibilities for localization, extraordinary transmission, metamaterials, hot-electron developments and new instrumentation (such as modern electron energy loss spectroscopy). The field has been good for surprises.

Certainly, hype is a problem in the field, but I cannot confirm if it occurs more or less than it does in other fields. The promises and perspectives we make in our grant applications have changed a little bit but they are pretty much along the same lines as ten years ago, which is positive. However, in terms of specific applications, many things did not work out.

For example, at the time when you were working on those waveguides, we all hoped that telecommunications applications were around the corner. There were European projects dedicated to this, and small spin-off companies for Pierre Berini (Canada) and Sergey Bozhevolnyi (Denmark), who were trying to commercialize long-range plasmon stripe waveguides. Those applications did not lead to real products, but I think that is the normal way of things. We were optimistic. If you are too pessimistic you can not go anywhere, but if you are always promising impossibilities it is the same thing. So we aim for something in between.

What bothers me more are the cases of 'story-telling'. Of course this is a rather subjective thing that is hard to define, but if you are in the field for long enough you recognize things being redone with different names and 'stories'. One example for me is some of the work on optical antennas. Some of this seems to be redoing previous work using 'antenna language', where I do not really see the increase of physical insight. It is sometimes — not in all cases — something known but retold in other terms. This is also related to the works discussing electronic circuit analogues of plasmonic systems. At first I was very enthusiastic, but down the road I am not sure it really helped many people.

What does the future hold for plasmonics?

There are a lot of nice things you can do with plasmons, but I have not seen, so far, that you can significantly improve a solar cell with them. I like the idea personally, but when you look at it more closely, usually you can do things slightly better with dielectrics. However, I am quite enthusiastic about photodetectors and colour filters. Existing colour- or polarization-sensitive diode arrays work via some interference layers that you have on the individual diodes — it is difficult to process. Hole arrays seem to be accessible in terms of transmission, and you can tune the spectral properties, just with one metal layer. Plasmonics has always surprised us, especially in the last two or three years, when we learned to use electron energy loss spectroscopy, cathodoluminescence and other new tools. Using electron probes for investigation will really open new possibilities to look closer into plasmonics, down to the 1-nm scale. Electronic transport, local heating, catalysis, quantum optics and, of course, metamaterials are all topics where plasmonics can be active for some time to come.

INTERVIEW BY DAVID PILE