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Observing the Unobservable. Quantum Interference of Complex Macromolecules

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In my laboratory I work on a Kapitza–Dirac–Talbot–Lau interferometer for large and complex molecules. In this interferometer we have demonstrated the quantum interference for the largest objects that have shown quantum interference so far—well, at least we claim it’s quantum interference. Those were molecules with a mass of more than 10,000 atomic mass units, which is about the mass of more than 10,000 hydrogen atoms. The interference pattern that we get looks like that shown in the inset of Fig. 1.

It is quite different from the patterns that we saw in the last talk. I will explain the reason for the difference in a second. This pattern is basically obtained by using an additional grating as a detection mask that is scanned over the molecular beam.

Our experimental setup is shown in Fig. 1. It is an interferometer that works with three gratings. The first grating is our source grating, which creates the coherence of our matter waves. We need the source grating because these matter waves are produced by simply heating a sample of the molecules using a very crude method, namely a ceramic cylinder around which we wrap some heating wire. They leave this oven through a slit and enter the vacuum chamber with a thermal velocity distribution, so they are everything but coherent. They never actually become coherent in the forward direction because we just cut out something like 20% of the velocity spread. But what we really need for seeing interference is spatial coherence, that is coherence transverse to the direction of the propagation
of the molecules. This coherence is obtained by putting the first grating in the way of the molecular beam, and each opening, each slit of the grating, now acts as something like a point source. After this grating, the matter wave with which we describe the center of mass motion of our molecules coherently illuminates a few nodes of the second grating. This second grating in our case is not a material grating anymore, but it is created by retroreflecting a laser from a mirror such that it forms a standing light wave.

![Fig. 1](image)

This sketch shows the main components of the setup of the Kapitza–Dirac–Talbot–Lau interferometer for matter waves. The molecules emanate from a crucible and form a molecular beam that passes three gratings and finally is ionized and detected. The inset shows the measured interference pattern, a sine-like modulation of the count rate that results when the third grating is scanned over the molecular beam (Source: Tüxen et al. 2010, 4145–4147).

So you see, in earlier times people diffracted light at matter; we now diffract matter at light. This works in the following way: the standing light field produces a periodic electromagnetic potential. In this electromagnetic potential the electrons are shifted inside the molecules. This induces a dipole moment in the molecule and this dipole moment then again interacts with the electromagnetic potential.

This interaction imprints a phase shift on the matter wave, induces in it a position-dependent shift of its momentum. As I already said, we use a third material mask of the same period to scan over the molecular beam, and behind this grating we ionize the molecules and count them in a quadrupole mass spectrometer.
Observing the Unobservable

You need a very good vacuum to see the interference effects. When the molecules interact with background gas on their way, you will lose your interference contrast. The actual setup is something like three meters long and is much emptier than many parts of the solar system, which contains a lot of dust and dirt. The reason why this pattern looks like such a nice sine curve is that we perform our experiments not in the far field, which was described in the talk before, but in the near field. Fig. 2 shows the transition between these two regimes, the near and the far field.

You see that behind these narrow openings of the diffraction grating the waves evolve in a very chaotic way. You cannot really solve analytically what is happening there. But from all this chaos a certain order arises when at certain distances the pattern of the diffraction mask is reproduced, and this distance is the so-called Talbot distance. Also, you can see that at half the distance the pattern is reproduced with twice the period and so on. The structure that evolves here is sometimes called a Talbot carpet. On the right hand picture of Fig. 2 you see how the near field transits into the far field; this is not a sharp transition and where it happens depends on how many slits the diffraction mask has. In each Talbot order the outermost maxima of the pattern evolve into the far field. The number of slits determines how often the grating mask is reproduced, that is, how many Talbot orders you will see.

So, we put our detector somewhere in the second Talbot order, where you see a reproduction of the diffraction grating, and that is why you see such a nice sine curve here—because the potential of the standing light wave is a sine in the first order. Of course when I came to this conference I asked myself is this a computer simulation already? This fitting a sine curve into our data? I would say it is, but I'm not sure. I'm not sure what a computer simulation is exactly. Well, one thing for sure that my predecessors on the
experiment did was asking the question about whether this diffraction pattern that we see really is a quantum diffraction pattern. Or is it just the result of classic ballistic diffraction, like of footballs hitting the goal post? They did a simulation where they compared how the visibility, the contrast of your interference pattern, would behave for different laser powers and the result are shown in Fig. 3.

![Fig. 3] Fringe visibility as function of diffraction laser power. Measured data compared to simulation for classical and quantum interference (Source: Hornberger et al. 2012, 157–173).

The blue line gives you the development of the visibility for a classical theory and the red line gives you the predictions of quantum theory; you can see that the experimental results agree quite well with quantum theory, but definitely do not agree with a classical approach.

However, although we heat up about half a gram of molecules in our oven and many thousands of molecules are flying through the grating at the same time, what we see is not interference of molecules with one another, but of each molecule with itself. We claim that it has to be interference of the molecule with itself because the molecules are very hot. They have many internal degrees of freedom, many hundred degrees of freedom. It’s very unlikely that two of the molecules are in the same state at the moment they are simultaneously passing through the grating. If they are not in the same state, they can be distinguished—and two distinguishable objects can not coherently interfere with each other. So what we see is the interference of molecules with themselves. But there is something really puzzling going on: interference should be something that only happens to waves. One has to be very careful to be clear that what we are looking at in our theoretical models is the center-of-mass wave function. It is a wave function that describes the motion of the center-of-mass of a really big object, and the wavelength is actually orders of magnitude smaller than the object. It doesn’t tell us what happens to the components of the object, and
we can interact with the object as if it was a complex particle with an inner structure. For example, we can measure the distribution of charges inside the molecule. Actually, when I told you that the light grating works because charges are shifted inside the molecules, I was using a particle picture to describe the diffraction of a wave. This is really weird to me, and it is also really weird to me that we can use the interference to probe the particle properties of the molecules, such as their electric polarizabilities or their permanent magnetic moment. These are properties that result from the internal structure of the molecules and that are not really part of my wave picture of these entities.

We can also do absorption spectroscopy in our interferometer—we send photons into the chamber, where they cross the molecular beam. When their wavelength is resonant with a transition in the molecules, they absorb the photon and get a kick to the side. While the matter wave is delocalized transverse to the direction of its center-of-mass motion, an absorption event takes place that is much more localized in the direction of this motion itself. In a way what happens in the experiment is something very strange, because we have a localized absorption of a photon by a molecule that is actually undergoing an interference process with itself. So it should be delocalized, and it is indeed delocalized in one direction and localized in the other direction.

References
Tüxen, Jens, Stefan Gerlich, Sandra Eibenberger, Markus Arndt, and Marcel Mayor. 2010. “Quantum interference distinguishes between constitutional isomers.” Chemical Communications 46 (23): 4145-4147.
Discussion with Lukas Mairhofer

Eric Winsberg: Just a quick question about the comment you made about how it's weird that you're treating the molecule as a wave function, but then it has all these internal degrees of freedom that matter. Is that different from when you use an electron? After all, you might just look at the spin of the electron, which is a very reduced representation of it in respect to the electron's degrees of freedom. Maybe it's made up of some...

Lukas Mairhofer: Well, you don't have this many degrees of freedom in an electron so it's easier, or think of photons.

EW: Let's try electrons, right? You could look at an inner structure of an electron.

LM: Supposedly an electron is a point-like particle that has no inner structure. Of course you can prepare atoms in different states but it's easy to prepare them in the same state. It's really, really hard to do that with large molecules.

EW: What is the molecule that you're looking at?

LM: Well our working horse molecule is the fullerene C60; it consists of 60 carbon atoms and looks like a football with its round shape and the structure made up of pentagons and hexagons. But we use many other molecules, some tailor-made by chemists, some just as they exist, for example in biological systems. Right now we're doing interference with vitamins A, K, H and D. We are trying to show interference with longer chains of peptides and proteins, in the future maybe with a viroid. So those are the molecules we are working on. They are large enough to be called Schrödinger's cats, definitely, yes—it's really hard to prepare two cats in the same state.

Stefan Zieme: I guess the size of the molecule— I mean, how big can they be? It's just a question of how good the vacuum is so can you make an estimate on how far you can go and if you can make an estimate about whether it converges? What is the boundary between classical and quantum?

LM: That's a very interesting question, and that of course is a question that also drives us because it is at the foundations of physics. First of all it's not only a question of the vacuum... your de Broglie wavelength, that is, the wavelength of your matter wave, scales inversely with
your mass, so your de Broglie wavelength becomes very small when your mass increases, and then to see the interference effect your interferometer needs to become very long. If you want to build an interferometer for a viroid, for the RNA strand without its protein shell, with the sources and the techniques that are available at the moment, it will be something like... each arm will be something like one or one and a half meters long. At the moment in our interferometer each arm is 10 centimeters long. Of course you need a good vacuum then. Also vibrations really become a problem when you have such a long interferometer. Even in the interferometer that I work on now you lose half of your contrast if your grating period is misaligned by half an Angström, that is half the radius of a hydrogen atom, for example because your laser wavelength has changed or something like that. So things like this are limiting you in a technical way. And then on the fundamental level, some theories claim that there is a limit on the size of the objects that you can show interference with. Because the question is, why do we not see quantum effects like interference in our everyday experience? Why does the world we live in seem to follow such a radically different physics? There are many approaches to explain this, and one is to claim that there is a spontaneous collapse of the wave function under its own gravity, for example. That would scale with the mass of the particle. Early spontaneous collapse models derived that you shouldn’t see interference above 2,000 atomic mass units. Then it was about 10,000 atomic mass units, now it’s about 100,000 atomic mass units. So there are some parameters you can tweak, but it seems that you cannot tweak them arbitrarily. At some point this model can be ruled out and this we try to test in our interference experiments.

EW: I mean in the Ghirardi–Rimini–Weber (GRW) theory, it depends on time. You can have an arbitrarily large thing not collapse, according to GRW, for a very short period of time.

LM: Yes. We look at it at reasonably long times, a few milliseconds.

Martin Warnke: I have a question because you yourself put up so many doubts and spoke of your puzzlement—my biggest puzzlement is having seen you with your young colleague in the laboratory, filling in that blue stuff at the left-hand side of the experimental system. Using a spoon, taking lumps of C60 atoms out of a box, putting them into the oven. Then you closed the apparatus and drew a very high vacuum. After that preparation in the real world with real and hard matter, then
in the world of an isolated apparatus you perform an experiment that you describe as one where matter waves interfere with themselves. The blue material from the beginning transforms, or should I say, trans-substantiates, into uncorporal waves. How do you do that in your mind? Say in one quarter of an hour you’re putting blue stuff into the left-hand side and after a few hours, when the vacuum is up again, you’re thinking of matter waves. How do you do that?

LM: I asked that myself for a very long time, until after a bottle of red wine I thought of myself in a space suit drifting through a dark universe without any point of orientation and without any interaction, without any stars around me. Completely blind, completely isolated. I thought I might well think of myself as being delocalized then. What is the meaning of being localized when there is no frame of reference? When there is nothing you can map your location to, if there is no interaction with your environment? I think even in our human minds, we could imagine being delocalized—or at least the concept of being localized would lose its meaning.

Hans-Jörg Rheinberger: I have a little problem with the probes that you’re using. So if you’re using that fullerene, it somehow makes sense. But if you think of a protein, that usually only exists with a lot of water molecules around it and so on and so forth. So what do you do to these molecules before you shoot them into the vacuum and what happens to them in the vacuum?

LM: Proteins are not very happy in a vacuum, that is true. The proteins unfold, so they spread out. But for example, the aim with the virus would of course be to show that it is still reproductive afterwards and it is still this half-living thing that it was before. It’s actually this transition between the gas phase and the in vivo environment that interests us so much. We try to attach water molecules to the protein in a controlled manner after we evaporate it, to see how that changes its behavior in the interferometer, its absorption of light and so on. To give you an example that is not a protein but that will make it clear why this is interesting, consider retinal, the molecule in your eye that triggers the visual process when it absorbs a photon. You know that you have different cells for blue light, for green light, and for red light. But the interesting thing is that it’s always the same retinal in these cells. The shift in the absorption line is only caused by the protein that it has bonded to. It would be really interesting for biologists and chemists to know where retinal absorbs when it is alone, when it is in
the gas phase, and nobody knows because you cannot resolve it with classic methods—only the sensitivity of our interference patterns will allow us to measure that.

Kristel Michielsen: Maybe I missed it, but how many molecules do you have in your interferometer?

LM: Our detection efficiency is lousy. We detect one in ten thousand to one in a million of the molecules arriving at the detector. When you run the interferometer, when you run a scan, you have something like 300, or okay, let's say you have something between 100 counts to 1,000 counts per second, but you can multiply this by a reasonably large number to get the actual number of molecules we have flying in there. The time of flight through the interferometer is a few milliseconds.

KM: So you have a bunch of molecules that goes at the same time?

LM: Yes, but they are distinguishable, they are not in the same state. So that at least in the quantum mechanical description you cannot make them interfere with one another.

Arianna Borelli: Of this question of the interference, because it was not very clear to me, what you meant that they cannot interfere if they are not in the same state, maybe you can make that clear, but now another question came to me. You speak always of waves, as far as I can tell, and never of fields. Of course if you would think of fields you would think there's this molecule field with different waves on it and then of course they might interfere with each other, waves in the same field—and now, moving into the mathematical world: if I think of these waves and waves in a field then they can all interfere. If you had to talk about fields, now speaking again in the mathematical world, would you say each of these waves is a different field or does that not make sense? As I said this is a former problem, but it's interesting for me to understand what you mean by interference and waves.

LM: The question of the field is very difficult for me because I have never seen a quantum field theory description for such huge molecules. I also have to admit that for this question I'm a little bit too much on the experimental side. As far as I see in the theory, I don't find an approach for a field theoretical model for what we do—just because the particles are too complicated. And yes, for the description of the center of mass motion you have a wave function.
AB: So you have a wave and, maybe I can put it in a more concrete way, these waves are waves in space and each particle has its own space? Its own space variables? Okay?

LM: Yes.

MW: May I quote Markus Arndt, the head of the group? When we talked to him he said there is actually no applicable theory for this situation and that we tried to measure what could not yet be calculated. Which is a very interesting point of view. Just a quote.

Anne Dippel: Hans, Kristel, what Lukas shows here right now, that’s something you could model, but then this is not a simulation. You can’t calculate it without a field theory.

LM: Yes, that’s great. Isn’t the simulation something you always can calculate?

MW: Maybe an analog simulation. I have just another question: You showed simulations but you didn’t name them. So the graphics you used, as far as I know, are from the Duisburg group simulating the near fields. It’s very peculiar for me. I know that your very highly esteemed colleagues in Duisburg are doing this, but why is this always something mute and invisible in your work? Why are there computer simulations that are of extreme importance for the Talbot carpet, which come, as far as I remember from the papers I’ve read, from the simulations they do in Duisburg. Could you describe the relationship between the experimental work and the computer simulations that were done beforehand, which you never talk about?

LM: Actually that’s a thing I really forgot, because at one point I thought, ‘Oh nice—I’ll put in this picture and then I’ll tell you that’s the result of a computer simulation.’ Especially this Talbot carpet; it’s a numerical simulation of one of my colleagues. But we have a very strong collaboration with the group in Duisburg that has been doing the theory for many years. One member of our group, who developed the theory for all of our interferometers, joined the Duisburg group after doing his PhD with us. So there are very strong links to them. For me...why I don’t talk about that work has two reasons. The first is that I don’t understand it completely and it’s their work. It’s hard for me to present it. The other thing is: for me, there is very much the question that I asked in the beginning. What is a simulation? Because what they are doing is of course, that they develop models. They do that together with us, and model what is going on in the interferometer. Then they
write the model as MATLAB code for it and then they basically do a fit on our results for free parameters and they get a lot of information about our interferometer. Is this a simulation where you simulate what would happen in an experiment? No—it’s a fit on existing data based on a model, and I don’t know if this is a simulation—I just don’t know it. It might well be that you call this a simulation; for me it is more a reconstruction of data.

Sonia Fizek: I actually wanted to ask you why can’t you just go digital? Why do you need to do it the way you do it, and now you’ve kind of answered that. Maybe the simulation, you could call it a simulation the minute you can change variables. So let’s say you have this problem with the length of the arm in a simulation: in a digital world you could just remove that variable and it is no longer there. So you kind of falsify things and maybe that’s when you can talk about simulating stuff that is not 100% a reflection of reality in your lab. Maybe they do it?

LM: I agree, if you start to think ahead about what is going to happen if you do this and this, that for me would be a simulation. Exactly.

Hans de Raedt: In this picture you showed a grating that looked perfect, but I assume—in your experiment it’s quite different?

LM: Since the grating has been in there for something like eight or nine years, I’m afraid it really is far from perfect nowadays.

HDR: Let’s say you get it from—I don’t know who makes it…

LM: Nobody makes it anymore, that’s the problem.

HDR: Let me rephrase it: when you first got it, what were the specifications of these—are these openings the same?

LM: The period of the material grating is 266 nanometers, the opening fraction is 40%, and all the openings are supposed to be the same; after eight years that might not be true anymore. But this material grating is not the diffraction grating that produces your interference pattern. So if we talk about the actual diffraction grating, we need to talk about the laser. This laser we can specify very, very well. We can measure its wavelength with femtometer precision and keep it stable to a few picometers. We can measure its power very accurately and we can look at the profile of the beam when it enters and when it leaves the chamber. This is all necessary because this is an incredibly important screw for us to tweak.
HDR: I understand that. So the grating that you call G1 and G3 is of course essential for what you get out—what goes in the interferometer and also what you detect. Not for the pattern but for the...

LM: It is important for creating good coherence, and if you don't create good coherence you don't see interference anymore. It is also actually critical that all slits should look more or less the same.

KM: In your picture you mentioned that one molecule is self-interfering?

LM: Yes, that's what we would claim.

KM: So if one molecule arises and if you are lucky because of the detection efficiency you see the spot, very localized. So now the next one comes. How is your picture at the end. Do you find stripes in an interference pattern?

LM: I would claim that interference is not something that you can ascribe to a single particle or a single wave. For me interference is an ensemble phenomenon. You cannot, you will never resolve the interference pattern of a single interfering entity. As you said, you need a lot of them to see the pattern and I don’t have a problem with this.

KM: In your picture you have self-interference but you need many, so how do you provide this?

LM: Well, you need many entities that have been interfering with themselves. You describe an ensemble of entities that have been interfering with themselves—with themselves because they cannot interfere with the others. The concept of this self-interference is that the center of mass wave function gets split by at least two slits of your grating or nodes of your standing light wave. That it is...

KM: In a way that's a wave description of the ensemble. Not of one. How do you do this with one?

LM: Well how do you distinguish between the ensemble description and the description of one entity? You cannot get the ensemble if you don't have many “ones” and you cannot have any description of “the one” if you don't measure the ensemble. For me it's not possible to get one without the other. If you give a description of the ensemble, you give a description of all the entities in the ensemble but you do not describe the properties of the individual entities. You will never see these wave properties if you only look at the individual entity or event.