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Electrons in doubt: The double-slit experiment on molecules

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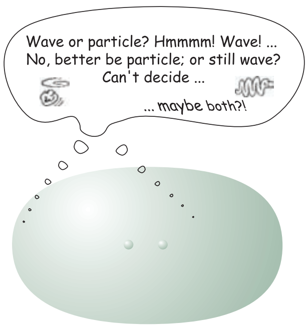


Electrons in doubt

The double-slit experiment, in particular performed with single electrons, is one of the key experiments of quantum mechanics. In 2002, the readers of the journal 'Physics World' even voted it to be the 'most beautiful experiment in physics' [1]: Although the electrons pass through the slits one at a time and appear as a single 'click' or speck on a screen, their sum still forms an interference pattern (Fig. 1). It seems as if each electron divides itself when passing through the double-slits, only to reunite again on the screen. However, if one slit is closed or if an observer determines which slit each electron passes through, the electrons behave like normal particles, that can only be at *one* point in space at a certain time, and the interference pattern disappears. In other words, depending on how the experiment is performed, the electron either appears to be at point A, or point B, or at *both* at the same time.

This apparent quandary is resolved by the particle-wave dualism and Bohr's complementary principle [2], which requires however that only *one* of the two forms of appearance, either particle *or* wave, can be observed at the same time. In recent times, scientists have questioned the validity of the complementary principle for photons as well as compound systems such as atoms [3] and fullerenes [4] as experiments have shown situations where matter appeared as particles and as waves at the same time – a gray area of complementarity. Would this gray area, or rather this area of coexistence, also be observable for structureless, non-compound particles like electrons?

We addressed this question in a 'molecular double-slit experiment' (Fig. 2) employing molecular nitrogen, N_2, as double-slit on atomic scale. We ionized the highly localized K-shell electrons in molecular nitrogen with polarized synchrotron radiation, knocking out one electron from each molecule. Because of the mirror symmetry of N_2, this electron belongs to both sides of the molecule in equal parts, and its emission is supposed to be coherent (i.e. phase-locked with phase 0 or π) from both atomic sides. The wave function ψ of the remaining ion is denoted with *g*, *gerade* (for phase 0), and *u*, *ungerade* (for phase π), in Fig. 2.

Until recently, it was impossible to separate the two wave functions concerning their angular dependence. However, this task has been achieved in the present experiment and it is now possible to observe each wave function separately. Both wave functions describe a state in which the electron exists at both sides simultaneously. Depending on the state, the wave function has either the same or the opposite sign on each side. The *gerade* case with equal signs corresponds to the classical double-slit experiment where the two sides of the molecule represent the two slits. Modern double-slit experiments with polarized photons can also generate the phase-shifted *ungerade* state. In that case, the interference pattern consists of so-called anti-fringes, where the dark and light areas are the inverse of the well-known interference fringes [5].

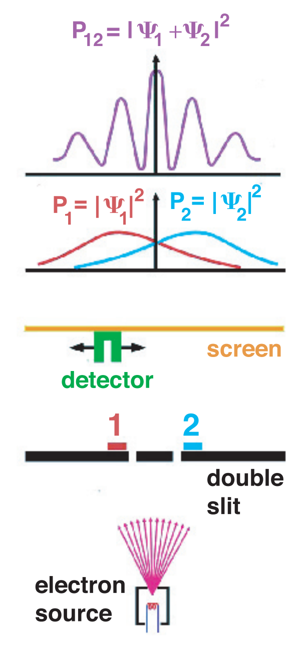


Fig. 1:
Double-slit experiment with single electrons. If one of the two slits is closed, a bell shaped shadow of the other slit is visible on a screen behind the slits. However, if both slits are open, an interference pattern of bright and dark stripes appears, from which no information can be inferred on the way the electron took to the screen.

References:

[1] R.P. Crease, Physics World Sep 2002; http://physicsweb.org/articles/world/15/9/2

[2] N. Bohr, Nature ***121****, 580 (1928).*

[3] S. Dürr et al., Nature ***395****, 33 (1998).*

[4] L. Hackermüller et al., Nature ***427****, 711 (2004).*

[5] S.P. Walborn et al., American Scientist ***91****, 336 (2003).*

[6] D. Rolles et al., Nature ***437****, 711 (2005).*

Acknowledgements:

Supported by the BMBF. D.R. also acknowledges support through the Feodor-Lynen program of the Alexander-von-Humboldt-Stiftung.

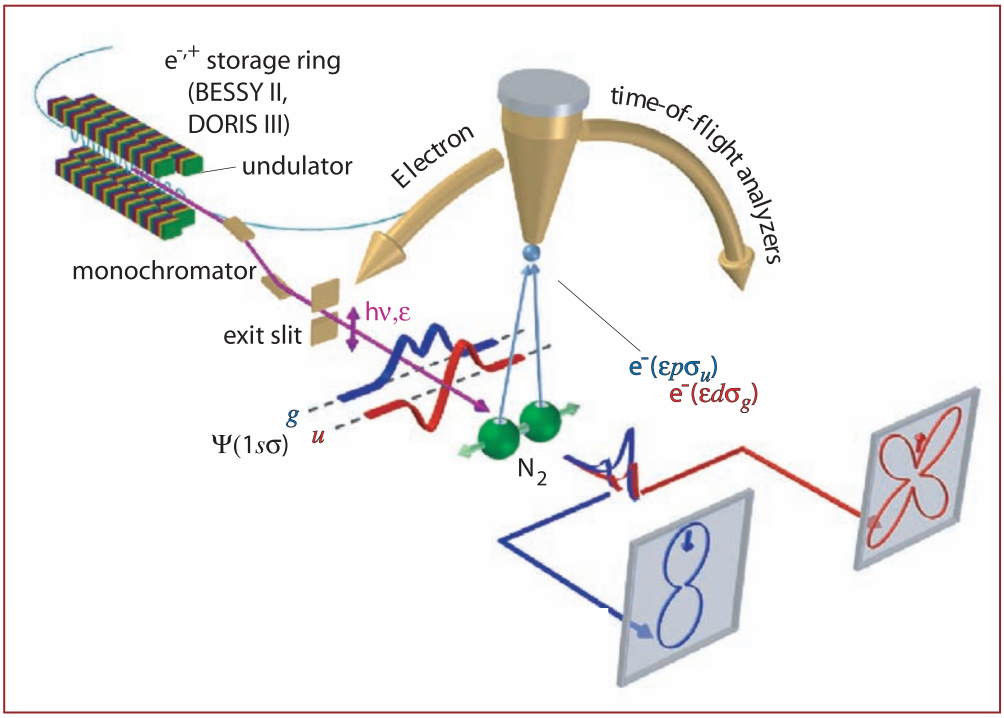
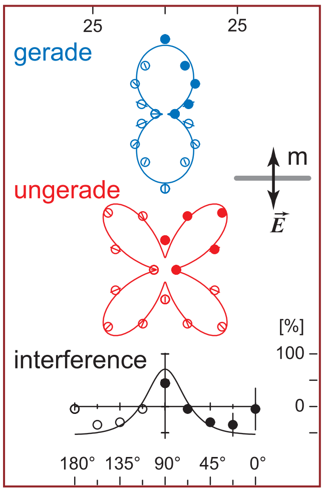


Fig. 2:
N$_2$-molecules are ionized with synchrotron radiation, and each molecule emits an electron from its inner-most shell. Depending on the phase-coupling inside the molecule, the electron is emitted with a slightly different energy, indicated by the red and blue spectral lines between the N$_2$-molecule and the 'screens' (right). Each one of those electron lines, called *gerade* ***(g) and*** *ungerade* ***(u), has a characteristic emission pattern, shown schematically on the corresponding 'screens'.***

In the molecular double-slit experiment, the molecular axes are statistically distributed in space. Therefore it is necessary to detect the electron in coincidence with the fragment ions created by the ionization. This allows measuring the angular distribution of the emitted electrons with respect to a fixed molecular axis. One can call this method a dynamical orientation of the molecules in the gas phase, where the orientation of each molecule at the instant of the electron emission is determined via the ionic fragmentation of the molecule. Fig. 3 shows the resulting electron angular distribution for the two states *g* and *u*.

In addition to demonstrating the coherent electron emission, an extra twist in the experiment also proved that breaking the molecular mirror symmetry leads to a loss of coherence. When repeating the experiment with a molecule made up of one lighter and one heavier isotope, in this case ^{14}N and ^{15}N, the electrons started to localize on one of the two, now distinguishable atoms. The electron distributions lost their strict parity-determining character and started to become more similar [6]. The shape of both angular distributions changed towards their common sum, as indicated by the arrows on the screens in Fig. 2.

Fig. 3:
Angular distribution of the *gerade* ***and*** *ungerade* ***electrons with respect to the molecular axis m for the 'perpendicular case' with the electric vector E of the ionizing radiation being perpendicular to m. The measurement shows the pure p- and d-character of the emitted electron waves. It is a direct proof for the existence of the non-local*** *gerade* ***and*** *ungerade* ***states in the molecule.***



This situation corresponds a *partial* marking of one of the slits in the double-slit experiment and proves that even for electrons, the complementary principle is not valid in its original form as exclusion principle. The electrons are wave and particle at the same time.

While these results certainly demonstrate very fundamental properties of quantum mechanics, they may also be of importance for more practical applications such as the study and control of 'artificial molecules', which consist of semiconductor quantum dots and which are envisioned as possible building blocks of future quantum computers.

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