Wave–particle duality is frequently the first topic students encounter in elementary quantum physics. Although this phenomenon has been demonstrated with photons, electrons, neutrons, and atoms, the dual quantum character of the famous double-slit experiment can be best explained with the largest and most classical objects, which are currently the fullerene molecules. The soccer-ball-shaped carbon cages \( C_{60} \) are large, massive, and appealing objects for which it is clear that they must behave like particles under ordinary circumstances. We present the results of a multislit diffraction experiment with such objects to demonstrate their wave nature. The experiment serves as the basis for a discussion of several quantum concepts such as coherence, randomness, complementarity, and wave–particle duality. In particular, the effect of longitudinal (spectral) coherence can be demonstrated by a direct comparison of interferograms obtained with a thermal beam and a velocity selected beam in close analogy to the usual two-slit experiments using light.

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I. INTRODUCTION

At the beginning of the 20th century several important discoveries were made leading to a set of mind-boggling questions and experiments that seemed to escape any answers based on classical, pre-quantum physics. The first were the discoveries\(^1\) that implied that optical radiation has to be composed of discrete energy packages that can be well localized in space and time. This localization was in marked contrast to the existing knowledge based on Maxwell’s theory which successfully represented light as electromagnetic waves. The second and complementary breakthrough was the theoretical result by de Broglie,\(^4\) and the experimental demonstration by Davisson and Germer\(^5\) that massive particles also propagate in a wave-like manner.

Both statements were stunning at the time that they were proposed and both keep us busy thinking even today because we generally associate the notion of point-like locality with a particle while we attribute spatial extension to a wave. The observation of both phenomena in one and the same experiment leads us also to the concept of delocalization, which goes beyond the simple concept of “being extended,” because single quantum objects seem to be able to simultaneously explore regions in space–time that cannot be explored by a single object in any classical way.

To illustrate the wave–particle duality we shall briefly recall the double-slit experiment as sketched in Fig. 1 because it is both one of the simplest and most general quantum experiments used in introductory quantum physics and is the prototype for our studies with molecules.

Let us first discuss an experiment that is usually performed in a ripple tank. If we excite surface waves in water and let them propagate through a small hole in a barrier (Fig. 1, left), we would observe a circular wavelet emerge behind the barrier in agreement with Huygens’ principle. If we now open a second hole in the barrier, we could create regions where the water remains completely still (Fig. 1, center). This phenomenon is simply explained by the fact that the surface waves superpose on each other and the wave minima can be filled by wave maxima at well-determined places. We call this phenomenon interference. It can only be easily observed if the disturbances in the two slits are synchronized with each other, which means that they have a well-defined and constant phase relation, and may therefore be regarded as being coherent with respect to each other.

For water the picture appears intuitive because the wave is composed of many particles, each interacting with its neighbors. But the experiment turns into the mind-boggler mentioned above if we repeat it with an ensemble of isolated objects—photons or even massive particles—which we send through the double-slit one by one.

We shall present experimental results with, at present, the most massive particles that exhibit wave properties. The results confirm that under appropriate circumstances we still obtain interference patterns, the shape of which can be predicted with certainty. However, it is important to note that in such investigations a single particle always gives a single click at one detector position only, and we have no means of calculating the position of this event in advance because, as far as we can tell, it is governed by chance.

Therefore, the double-slit experiment with single particles leads us to the following questions: How can a single particle, which we observe both in the source and in the detector as being well-localized and much smaller than a single opening in the barrier, acquire information about the state (open/closed) of a very remote opening, if it were considered to pass only one through the openings? Why can’t we track the particle position without destroying its wave nature? How can we understand the emergence of a well-defined interference pattern in contrast to the random hitting point of the single object if none of the particles can interact with the rest of the ensemble in any way that we know?\(^6\)

We thus find many fundamental quantum concepts in the context of double-slit interferometry. First, we find the complementarity between our knowledge about the particle’s position and the visibility of the interferogram. If we open one slit only, the particle must pass this opening and the interference pattern must disappear. Perfect interference contrast can be obtained only if we open the second slit and if we exclude all possibilities of detecting, even in principle, the path the object has taken. The wave–particle duality states that the description of one and the same physical ob-
mechanics: the double-slit experiment is at the heart of quantum defined position. In contrast to the property of having a well-
derminism. The pattern on the screen is well determined for the ensemble, but the detection point of a single object is also find the duality between objective randomness and de-
基本信息内容的量子系统本身是不足的，以至于不允许这样的描述，即使是原则。然而，相似
to light quanta in a laser beam, the atoms in a Bose–Einstein
decompose are kept sufficiently apart to keep their interact-
weak. Therefore, in spite of the large coherence length, the interfering object is still of small mass and complexity.

Even experiments demonstrating interference between two Bose–Einstein condensates can be viewed as a double-slit experi-
ent with many individual atoms, as witnessed also by the fact that to explain the fringe spacing the de Broglie
length corresponding to the individual atom rather than a wavelength using the total mass of the condensate is used.

Different questions and new experimental challenges arise if we study particles in the almost opposite parameter regime
where the interaction among the particles is much stronger. Covalently bound atoms form a new entity, a molecule or
cluster, and the de Broglie wavelength of this system is defined by the total mass of all the atoms and by the center-of-
mass velocity of the bound system. In the following we shall focus on these complex objects.

The very first demonstration of molecule interference dates back to the days of Estermann and Stern in 1930, who
demonstrated experimentally diffraction of H₂ at a LiF crystal surface. Further experiments with diatomic molecules had to
await progress and interest in atom optics. A Ramsey-Borde interferometer was already realized for the iodine
dimer in 1994 and was recently used for K₂. Similarly, a Mach–Zehnder interferometer was demonstrated for Na₂.
The near-field analog to the Mach–Zehnder interferometer, a Talbot–Lau interferometer, was recently applied to experi-
ments with Li₂. Diffraction at nanofabricated gratings also turned out to be the most effective way to prove the exis-
tence of the weakly bound helium dimer and to measure its binding energy.

Based on these historical achievements we ask how far we might be able to extend such quantum experiments and for
what kind of objects we might still be able to show the wave–particle duality. Recently, a new set of experiments
exceeding the mass and complexity of the previously used objects by about an order of magnitude has been developed in
our laboratory. These experiments with the fullerene molecule C₆₀ will be described in Sec. II.

II. THE C₆₀ EXPERIMENT

The cage-like carbon molecules earned their names “fullerenes” and “buckminsterfullerenes” because of their close resemblance to geodesic structures that were first discussed by Leonardo da Vinci and implemented in buildings...
in the United States by the architect Buckminster Fuller. This new modification of pure carbon was discovered in 1985 by Kroto et al. and shown to be particularly stable and abundant when exactly 60 carbon atoms are arranged in one molecule to form the smallest natural soccer ball we know, the buckyball, as shown in Fig. 2.

Fullerenes are appealing candidates because a successful quantum experiment with them would be regarded as an important step toward the realm of our macroscopic world: Many of the known physical properties of buckyballs are more closely related to a chunk of hot solid material than to the cold atoms that have so far been used in matter wave interference. The existence of collective many-particle states like plasmons and excitons, the rich variety of vibrational and rotational modes as well as the concept of an internal molecular temperature are only some of the clear indicators of the multiparticle composition of the fullerenes. And we might wonder whether this internal complexity could spoil the quantum wave behavior of the center of mass motion.

To answer this question, we have set up a new experiment as shown in Fig. 3. It resembles very much the standard Young’s double-slit experiment. Like its historical counterpart, our setup also consists of four main parts: the source, the collimation, the diffraction grating, and the detector.

A. The source

To bring the buckyballs into the gas phase, fullerene powder is sublimated in a ceramic oven at a temperature of about 900 K. The vapor pressure is then sufficient to eject molecules one by one through a small slit in the oven. The molecules have a most probable velocity $v_{mp}$ of about 200 m/s and a nearly thermal velocity spread of $\Delta v/v_{mp} = 60\%$. Here $\Delta v$ is the full width of the distribution at half height.

To calculate the expected diffraction angles, we first need to know the de Broglie wavelength which is uniquely determined by the momentum of the molecule

$$\lambda = \frac{h}{mv}, \quad (1)$$

where $h$ is Planck’s constant. Accordingly, for a $C_{60}$ fullerene with a mass of $m = 1.2 \times 10^{-24}$ kg and a velocity of $v = 200$ m/s, we find a wavelength of $\lambda = 2.8$ pm.

B. The diffractive element

Because the de Broglie wavelength is about five orders of magnitude smaller than any realistic free-standing mechanical structure, we expect the characteristic size of the interference phenomena to be small. A sophisticated machinery is therefore necessary to actually show them. As the diffracting element we used a free-standing silicon nitride grating with a nominal grating constant of $d = 100$ nm, slit openings of $s = 55 \pm 5$ nm and thickness of only 200 nm along the beam trajectory. These gratings are at the cutting edge of current technology and only a few specialists worldwide can actually make them.

We can now calculate the deflection angle to the first diffraction order in the small angle approximation as the ratio of the wavelength and the grating constant,

$$\theta = \frac{\lambda}{d} = \frac{2.8 \times 10^{-12} \text{m}}{10^{-7} \text{m}} = 28 \mu\text{rad}. \quad (2)$$

In elementary textbooks Eq. (2) is usually derived using Fig. 4 and noting that the first constructive interference occurs when the difference between two neighboring paths is equal to one de Broglie wavelength. Because our detector is placed at 1.2 m downstream from the grating, the separation between the interference peaks at the detector amounts then to only $L \times \theta = 1.2 \text{m} \times 28 \mu\text{rad} = 34 \mu\text{m}$.

![Fig. 2. The fullerene molecule $C_{60}$, consisting of 60 carbon atoms arranged in a truncated icosahedral shape, is the smallest known natural soccer ball.](image)

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![Fig. 3. Setup of the diffraction experiment. Fullere molecules are sublimated in the oven at 900 K. The spectral coherence can be improved using a mechanical velocity selector. Two collimating slits improve the spatial coherence and limit the angular spread of the beam to smaller than the expected diffraction angle. A SiN grating with a 100 nm period and 50 nm openings is used to diffract the incident molecular waves. The molecular far-field distribution is observed using a scanning laser-ionization detector.](image)

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![Fig. 4. Textbook approach to double-slit diffraction. First-order interference maxima of a monochromatic wave are caused by constructive interference of the wavelets that emerge from two neighboring slits. The corresponding path length difference between the two paths is equal to the de Broglie wavelength. Higher order interference will be spoiled by the limited longitudinal coherence in a thermal source. Velocity selection in our experiments increases the longitudinal coherence length by more than a factor of 3 and therefore permits the observation of higher order interference fringes.](image)

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C. The detector

The small spacing between the interference orders requires a high spatial resolution of the molecular detector. For the fullerenes we have implemented a novel detector that surpasses most other schemes in detection efficiency, spatial resolution, and simplicity.

A continuous-wave green laser beam (argon ion laser, all lines) with a full power of 25 W is focused to the beam width of only 4 μm. As shown in Fig. 3, the laser beam is orthogonal to the molecular beam. All molecules that pass the laser beam at or very close to the focus are heated to an internal temperature in excess of 3000 K and ionize. The positive fulleren ions are then accelerated toward an electrode at 10 kV where they induce the emission of electrons. The electrons in turn are again multiplied and the charge pulses are subsequently counted. The overall molecule detection efficiency is about 10% and thus about two orders of magnitude higher than for example, electron beam bombardment ionization as used in many mass spectrometers. We find that among all gases in our vacuum chamber, the laser detector is only sensitive to fullerenes, due to the particular level scheme and high stability against fragmentation. Because of the tight focusing of the laser beam, the effective width of our detector amounts to only ~8 μm, which is sufficient to resolve the individual diffraction orders. To record a diffraction pattern, we scan the laser across the molecular beam in steps of 2 μm. The interferograms shown below represent molecule counts as a function of the transverse laser position.

D. Coherence considerations

Let us now turn to the coherence properties of our molecular beam. In general, coherence means that there is a fixed and well-defined phase relation in space and time between two or more wave fronts.

The spatial (transverse) coherence of our source is almost negligible right after the oven. Inside the source, the coherence width is actually only of the order of the thermal de Broglie wavelength. As is true in general for extended sources with uncorrelated emitters, the visibility is then reduced by the fact that the many partial interferometers—each starting at one point in the source and forming two trajectories through the double-slit toward a point in the detector—acquire different phase differences along their path to a given spot on the screen.

After the oven, we therefore need to enlarge the spatial coherence width by about five orders of magnitude in order to illuminate at least two neighboring slits coherently. Luckily, the spatial coherence is essentially determined by the geometry of the experiment and grows linearly with increasing distance from the source and with decreasing size of the first collimation slit. This general rule for the influence of collimating elements on transverse coherence is commonly known as the van Cittert–Zernike theorem: the spatial coherence function can be derived from diffraction curves which are determined by the apertures along the molecular beam. The limiting element in our case is the first collimation slit.

Obviously the gain in coherence has to be paid for by a dramatic drop in the count rate because the signal decreases quadratically with the distance from the source and linearly with the size of the slit. Although the first collimating slit alone already provides coherence, we still have to introduce a second collimating slit—in our case also 7 μm wide and about 1 m downstream from the first slit. The reason for this is the requirement that the collimated beam width needs to be significantly smaller than the separation between the diffraction orders behind the grating in order to clearly resolve the diffraction peaks.

The spectral coherence of the source also enters because molecules with different velocities and therefore different wavelengths follow different diffraction angles. And because the detector records the sum of the correspondingly stretched or compressed diffraction pictures, the interference pattern would be washed out. And in contrast to the spatial contribution, there is no gain in longitudinal (spectral) coherence during free flight. This is due to the fact that different velocity classes will evolve differently. In a pulsed beam experiment we would therefore observe a chirped packet, that is, a wave packet with short wavelengths in the pulse lead and long wavelengths in its tail. And even though the packet would spread out in the course of its evolution, the coherence would not grow due to the internal rearrangement.

Although even in pulsed experiments the spectral coherence does not improve during propagation because of the internal restructuring of the wave packet, the picture of a wave packet is problematic for the description of a continuous source. It is unfounded because the wave packet picture implies a well-defined internal phase structure. More specifically, a wave packet is characterized by a well-defined phase relation between different Fourier components of the beam. Yet such a relation can only be imposed by a suitable preparation. In our case that would imply a well-defined time at which the wave packet starts. This is not provided in our experiment, and the beam can be regarded only as a statistical, and therefore incoherent, mixture of the various momenta. Nevertheless, the beam can operationally be characterized by a coherence length, which is the length that measures the falloff of the interference visibility when the difference between two interfering paths increases. The longitudinal coherence length is given by

\[ L_c = \frac{\lambda^2}{\Delta \lambda} \]

For our thermal beam with \( \Delta v/v \sim 0.6 \) we find \( L_c \sim 1.7 \lambda \), which is just enough to guarantee the existence of the first-order interference fringes. We shall later discuss the

Fig. 5. Velocity distribution of the C_{20} molecules for a thermal and a velocity selected beam. The thermal beam (gray curve) is centered around \( \bar{v} = 200 \) m/s and has a width of \( \Delta v/v \sim 0.6 \), while the selected beam (black curve) is centered around \( \bar{v} = 117 \) m/s with a width of \( \Delta v/v \sim 0.17 \). We therefore expect the velocity selected interference pattern to be expanded by 70% on the screen and to show at least three times \( (\approx 0.6)0.17 \) as many interference orders as the unselected pattern.
improvement of the spectral purity using a velocity filter (see Figs. 3 and 5), thereby also improving the wavelength distribution.

Figure 6 shows a typical fullerene diffraction pattern with a thermal beam. We can clearly discern the first interference orders on both sides of the central peak. But the limited coherence is reflected by the fact that we cannot see any second or higher order peaks in the interferogram of Fig. 6.

To see more fringes we have to increase the coherence length and therefore decrease the velocity spread. For this purpose we have employed a mechanical velocity selector, as shown after the oven in Fig. 3. It consists of four slotted disks that rotate around a common axis. The first disk chops the fullerene beam and only those molecules are transmitted that traverse the distance from one disk to the next in the same time that the disks rotate from one open slot to the next. Although two disks would suffice for this purpose, the additional disks decrease the velocity spread even further and help eliminate velocity sidebands. By varying the rotation frequency of the selector, the desired velocity class of the transmitted molecules can be adjusted. To measure the time of flight distribution we chopped the fullerene beam with the chopper right behind the source (see Fig. 3). The selection is of course accompanied by a significant loss in count rate, but we can still retain about 7% of the unselected molecules.

In Fig. 5 both the thermal and the selected velocity distributions are shown. In contrast to the width of the thermal spectrum, amounting to $\Delta v/v = 60\%$, we are able to reduce this number to only 17% with the selector. The increase in longitudinal coherence by a factor of more than 3 allows for the observation of diffraction peaks up to at least the second and possibly the third order, as can be seen in Fig. 7.

It should also be pointed out that by using the velocity selector, we can now choose a slow mean velocity centered about 120 m/s, which corresponds to a de Broglie wavelength of 4.6 pm. It is obvious that this increase in wavelength results in a wider separation of the diffraction peaks, which can be seen by comparing Figs. 6 and 7.

In principle, the diffraction patterns can be understood quantitatively within the Fraunhofer approximation of Kirchhoff’s diffraction theory as it can be found in any optics textbook. However, Fraunhofer’s diffraction theory in the context of optics misses an important point that becomes evident in our experiments with matter waves and material gratings: the attractive interaction between molecule and wall results in an additional phase of the molecular wave function after the passage of the molecule through the slits. Although the details of the calculations are somewhat involved, it suffices here to say that the qualitative effect of this attractive force can be understood as a narrowing of the real slit width toward an effective slit width. For our fullerene molecules the reduction can be as big as 20 nm for the unselected molecular beam and almost 30 nm for the velocity selected beam. The stronger effect on slower molecules can be understood by the longer and therefore more influential interaction between the molecules and the wall. However, a complete description would need to take into account the correct shape of the complex (imaginary and real) transmission function, which implies the position-dependent modulation of both the molecular amplitude and phase.

The full lines in Figs. 6 and 7 are fits of our data to this modified Kirchhoff–Fresnel theory. To obtain such a good fit we also have to take into account an enhanced contribution in the zeroth order which we attribute to mechanical defects (holes) of the grating which are significantly larger than the grating period.

III. CONCLUDING REMARKS

A. Single particle interferometry

It is important to note that the interference pattern is built up from single, separate particles. There is no interference between two or more particles during their evolution in the apparatus. Single particle interference is evidenced in our case by two independent arguments.

The first argument is based on the spatial separation between the molecules. The molecular flux at an average speed of 200 m/s is $\sim 3 \times 10^6$ cm$^{-2}$ s$^{-1}$ at the plane of the detector. This flux corresponds to an average molecular density of $1.7 \times 10^{13}$ m$^{-3}$ or an average molecular distance of 200 $\mu$m. This is three orders of magnitude wider than any realistic range of molecular (van der Waals) forces, which are typically confined to several 100 nm.

The second argument is based on the fact that interference occurs only between indistinguishable states. However, all molecules may be regarded as being in different states. There
are 174 different vibrational modes and the rotational modes can be populated at different energies. The chance of having two subsequent molecules in exactly the same state of all internal modes is vanishingly small. Therefore, interference in our experiments really is a single particle phenomenon!

B. Coherence and which-path information

We might believe that coherence experiments could be spoiled by transitions between the many thermally excited states. Obviously, this is not the case, as has been shown by our experiments. But why is this so? No matter what we do, we can only observe one of these qualities in its ideal form at any given time. If we tried to locate the particle during its passage through one of the two slits, say by blocking one of the openings, the interference pattern would disappear. This rule still holds if we do not block the slit, but manage to obtain which-path information for example via photons scattered or emitted by the molecules. Sufficiently complex molecules, in contrast to the electrons, neutrons, and atoms used so far, may actually emit radiation without any external excitation, because they have stored enough thermal energy when leaving the oven. According to Bohr’s rule, the interference pattern must then disappear if the molecules emit a photon with a sufficiently short wavelength which enables the experimenter to measure the location of the emitting molecule with sufficient precision. According to Abbé’s theory of the microscope, the photon should have a wavelength shorter than twice the distance between the two slits.

What actually saves the experiment is the weakness of the interaction. The wavelength of the most probably emitted photons is about a factor of 100 larger than the separation between two neighboring slits. And the number of light quanta that actually leak into the environment is still suffi-

cient. The wavelength of the most probably emitted photon with a sufficiently short wavelength which enables the experimenter to measure the location of the emitting molecule with sufficient precision. According to Bohr’s rule, the interference pattern must then disappear if the molecules emit a photon with a sufficiently short wavelength which enables the experimenter to measure the location of the emitting molecule with sufficient precision. According to Abbé’s theory of the microscope, the photon should have a wavelength shorter than twice the distance between the two slits.

What actually saves the experiment is the weakness of the interaction. The wavelength of the most probably emitted photons is about a factor of 100 larger than the separation between two neighboring slits. And the number of light quanta that actually leak into the environment is still sufficiently small—of the order of one, up to potentially a few photons—and cannot disturb the interference measurably. Therefore, even if the fullerene molecule emits a few photons on its path from the source to the detector, these photons cannot yet be used to determine the path taken by the molecule. In other words, the photon state and the molecule state are not (or only very slightly) entangled because the two possible photon emission states from either path largely overlap. In a sense we may say that the fullerene has no “memory” along which path the emission occurred.

C. Conclusion

Quantum phenomena become increasingly important and the limit to which we may be able to confirm all quantum principles experimentally is still an open question. The discussion of our fullerene experiments lets us demonstrate the basic wave–particle duality for the most massive, most complex, and most “classical” single object so far. Many of the concepts that we teach our students can be illustrated simply. For instance, the notion of coherence length has a rather intuitive meaning when we compare the spectral width of the source and the number of observed interference fringes.

ACKNOWLEDGMENTS

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15A student, having been taught some elementary statistical physics, would be tempted to answer the first one by analogy with the Galton board. Also there, the trajectory of the single particle appears to be undetermined and still the count rate in each channel would be in agreement with the deterministic laws of classical statistics. But we should note, first of all, that the apparent randomness in this example is only due to the ill-defined initial conditions when we release the ball. Second, if we were to perform such a Galton experiment with two different starting positions, the probability distributions of these experiments would simply add up and interference would never be observed.
35C. Lisdat, M. Frank, H. Knockel, M.-L. Almazor, and E. Tiemann, “Re-


33 It is interesting to compare the de Broglie wavelength of the fullerene with its actual size: The buckyball has a diameter of about 1 nm, which is 350 times larger than its de Broglie wavelength. Our interference experiments clearly show that the concept of the de Broglie wavelength is not merely academic for objects with dimensions much larger than their wavelengths but can actually be demonstrated.

34 Our gratings were provided by Professor Henry Smith and Dr. Tim Savas of MIT.


38 E. Hecht, Optics (Addison-Wesley, Reading, MA, 2002), 4th ed.


40 The van der Waals interaction scales like $r^{-3}$ with the distance $r$ between molecule and grating walls. For C$_{60}$ the scaling even starts to change into a $r^{-4}$ behavior at distances beyond 20 nm, due to the finite (retarded) signaling time between the molecule and its mirror image; see also H. B. G. Casimir and D. Polder, “The influence of retardation on the London-van der Waals forces,” Phys. Rev. 73, 360–372 (1948).


Erratum: “Quantum interference experiments with large molecules”

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In Fig. 3 the correct distance between grating and detector was \( L = 1.20 \pm 0.01 \) m as also stated in the text.

In the caption to Fig. 7 the mean velocity in the experiment of Fig. 7 was experimentally determined to be \( \bar{v} = 136 \pm 3 \) m/s instead of \( \bar{v} = 117 \) m/s as stated in the caption for Fig. 7. Note, that the mean velocity in Fig. 7 was selected to be higher than that of Fig. 5 in order to utilize a more intense part of the thermal velocity distribution.

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Erratum: “One century later: Remarks on the Barnett experiment”
[Am. J. Phys. 71 (6), 558–561 (2003)]

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The derivation in Eqs. (4)–(8) follows closely that found in “Vacuum Electrodynamics on a Merry-Go-Round” by John Ise and Jack Uretsky [Am. J. Phys. 26, 431–435 (1958)]. I thank Jack Uretsky for bringing this reference to my attention.

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