Edward S. Fry

Statement
and
Readings

Determinism, Einstein, and Quantum Mechanics

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Abstract

Einstein believed that quantum mechanics was an incomplete theory. In that context, he is famously quoted from a letter he wrote to Max Born in 1944: "You believe in God playing dice and I in perfect laws in the world of things existing as real objects. . ." The conceptual foundations of quantum mechanics will be discussed, and the decades long history of this contentious problem will be reviewed. A version of the breakthrough analysis by John Bell in 1964 that made it possible to experimentally test these heretofore philosophical arguments will be presented. Results of the experimental tests of the Bell inequalities and their present status will be discussed. Finally, a loophole free test of the Bell inequalities will be described. It is an experimental implementation of Bohm's version of the EPR argument using spin one-half fermions.

ELECTRON AND ATOMIC PHYSICS

The study of atoms and molecules has been at the heart of 20th-century physics. Higher accelerator energies have permitted the exploration of matter at ever smaller scales—the nucleus, the proton, and now quarks—but a careful analysis of atomic and molecular dynamics is still essential.

Atomic research has been revitalized by the advent of new experimental techniques. The use of lasers is widespread, particularly high-powered solid-state lasers, tunable dye lasers, and lasers at ultraviolet wavelengths. New accelerators dedicated to the product of synchrotron radiation are providing high-powered beams over a wide range of wavelengths. Computers and new detectors have streamlined data taking.

Bell's Inequality and Experimental Tests of Quantum Mechanics

Recent atomic physics experiments have made a definitive contribution to our understanding and interpretation of quantum mechanics. It is generally accepted that quantum mechanics makes statistical predictions that are in excellent agreement with experimental data. The interpretation of quantum mechanics for single microscopic events has, however, been the subject of widespread controversy. The focal point for this controversy in a classic paper by Einstein, Podolsky, and Rosen[1] in which they put forth the proposition that quantum mechanics is an incomplete theory. To complete the theory, additional variables would presumably be required, and the term "hidden variables" was eventually coined for them. These hidden variables would enable one to make precise, deterministic predictions at the microscopic level. For example, if a vertically polarized light beam is incident on a linear polarizer whose transmission direction is at 45° to the vertical, then quantum mechanics can only tell us that the probability each photon will be transmitted is 50%; however, if one knew the values of these proposed hidden variables, then for each photon one could predict with certainty (100% probability) whether or not it would be transmitted.

A milestone in the interpretation of quantum mechanics was the proof in 1965 by J. S. Bell[2] that no theory incorporating hidden variables and satisfying a physically reasonable condition of locality (a "local hidden variable theory") could reproduce all the statistical predictions of quantum mechanics. The proof involves studies of correlated systems and takes the form of an inequality which must be satisfied by the statistical results of any local hidden variable theory, but which may be violated in some situations by the statistical predictions of quantum mechanics.

The locality condition simply states that the effects produced by an analyzer or detector should be independent of the settings of other spatially separated analyzers and detectors. Physically, the Bell inequalities tell us that in any local hidden variable theory there is a limit on the strength of the correlations that may be observed in an experiment; in contrast, quantum mechanics predicts very strong correlations that may exceed that limit.

Bell's inequalities made it possible to test the validity of the entire class of local hidden variable theories by performing experiments in which the quantum mechanical predications violate them.[3,4] At present, the most definitive experiments are those involving observation of polarization correlations between two photons in an atomic cascade. The consensus of these experiments is that any local hidden variable theory is inconsistent with nature. The first of these experiments was performed in 1972 by Freedman and Clauser[5] using a cascade in calcium. The initial state of their cascade had zero total angular momentum ($J = 0$) and the intermediate state was very short lived (5 nsec). In 1976, Fry and Thompson[6] completed an experiment on ^{202}Hg with an initial state of total angular momentum $J = 1$ and a relatively long-lived intermediate state (120 nsec). They obtained a dramatic relative improvement in the available signal using a laser excitation scheme. This was especially important for examining systematics, which almost invariably weaken the correlation and lead to results satisfying the inequalities.

In the last two years, the original Freedman and Clauser experiment has been repeated by a group in France, but with several important variations and improvements.[7–9] First, they used a two-photon laser excitation scheme that dramatically improved again the signal-to-noise ratio. Second, they tested the inequality for various source-polarizer separations up to 6.5 m. Third, they did an experiment with two-channel linear polarizers (i.e., both orthogonal linear polarization signals were observed at each cascade wavelength). This enabled them to obtain the strongest violation of a Bell inequality ever observed. Fourth, they performed an experiment using time-varying polarizers in which the effective polarizer orientation is chosen in a time less than that required for a light signal to travel between the two analyzer systems for the two cascade wavelengths. This experiment did not rigorously satisfy Einstein locality, since the choice was made quasiperiodically rather than randomly; nevertheless, it provided an important step beyond the fixed polarizers of all previous experiments. In all the experiments performed by the French group, excellent agreement with quantum mechanical predictions and clear violation of the Bell inequalities was observed.

Small loopholes still exist, but the overwhelming evidence provided by these atomic cascade experiments stands against any theory that would supplement quantum mechanics with hidden variables and still retain the physically very reasonable condition of locality.

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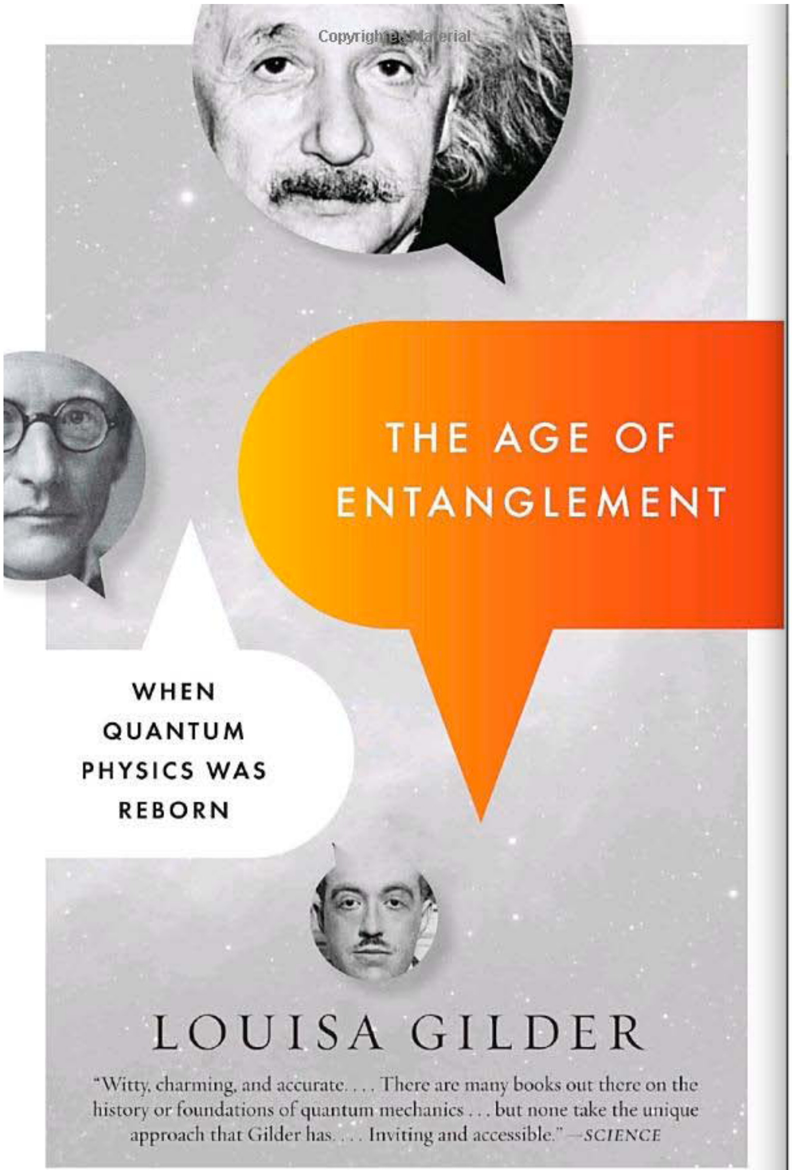

THE AGE OF ENTANGLEMENT

WHEN QUANTUM PHYSICS WAS REBORN

LOUISA GILDER

"Witty, charming, and accurate. . . . There are many books out there on the history or foundations of quantum mechanics . . . but none take the unique approach that Gilder has. . . . Inviting and accessible." —*SCIENCE*

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In Which the Settings Are Changed

1975–1982

Alain Aspect

HANGING NEXT TO THE DOOR of Bell's office at CERN was a poster of a long-necked Modigliani lady in a hat; her eyes and the eyes of Bell himself watched the twenty-seven-year-old Alain Aspect, a genial, mustachioed graduate student talking eagerly about boxes of water.

It was early in 1975, and Aspect had just returned to Europe from a three-year stint of French "national service," teaching in Cameroon. Soon after his return, he had suffered what he described as a *coup de foudre.* "In October 1974," he remembered, "I read John Bell's famous paper 'On the Einstein-Podolsky-Rosen Paradox,' and it was love at first sight. This was the most exciting subject I could dream of." Immediately he decided to make Bell's theorem the subject of his Ph.D. thesis at his alma mater, the University of Paris-South, in Orsay.

Meanwhile, Clauser was trying to get a job. "I must have applied to at least a dozen different places, and at all of them I was totally rejected." Universities were uneasy about hiring a professor who would encourage the next generation to question the foundations of quantum theory. Finally Clauser found an opening at the Lawrence Livermore National Laboratory, in the hills east of Oakland, researching plasmas (David Bohm's first love).

"I don't know anything at all about plasma physics," Clauser announced at his job interview. "But I do know a lot about doing experimental physics. I'm a very talented experimental physicist."

"You can learn plasma physics," was the reply. He was hired in 1976 and stayed there for a decade. At Livermore, his self-proclaimed skills as an experimentalist were well used. But almost wasted was an equal, though unproclaimed, skill. Clauser had the gift—rare even in the teaching profession—of explaining complicated subjects to students clearly, vividly, and patiently. The university career where he could have combined these skills did not materialize in the three decades since he first applied for such a job.

"Back in the sixties and seventies, reputable physicists did not ask questions about quantum mechanics," explained Fry in 2000. "I think that Clauser took the brunt of this attitude—in part, I believe, because he was actually *doing* the experiment, not just talking about the theory."

Fry himself had better luck with academia. In the midst of performing his experiment, he was granted tenure. Thirty years later, by then the head of the physics department at Texas A&M, he learned that this open-minded institutional decision was thanks to an intervention from Frank Pipkin, Holt's adviser at Harvard. Realizing that the tenure committee was about to reject the Bell experimenter, one of Fry's friends asked Pipkin to come to College Station, Texas.

"If you had sent me just Ed's file to look at, I would have rejected him very quickly," Pipkin told the committee. "However, after spending a day in his lab I can tell you that this guy is a winner and I would bet on his success." Pipkin's renown in atomic physics won over the skeptical committee.

Bell himself was acutely aware of the stigma attached to the experiments his work had inspired, but thus far Aspect was not. Before heading to West Africa in 1972, Aspect remembered, "I had a quite good education in classical physics, and I knew my education in quantum physics was extremely *bad*." The classes he had taken on the subject comprised equation-solving with little discussion of physical meaning, let alone inculcation of any stigmas.

So for his three equatorial years in Cameroon, Aspect taught himself quantum mechanics, using a recent textbook by the great French physicist Claude Cohen-Tannoudji. This book had two strengths: "First, it is real physics," said Aspect. "Second, it is neutral with respect to foundations. No brainwashing, no 'Bohr solved all of that.' " As a result, "I was able to solve the equations but nobody had washed my brain.

"I was totally convinced by Einstein and Bell," he said. But what experiment to do? In rereading Bell's 1964 paper, Aspect realized that its last lines told him "there was still an important test to be done."

He raced to Geneva to tell Bell his idea.

Bell had ended his paper on a cautionary note. If there was enough time for a light-speed signal to correlate the particles, then entanglement would lose much of its mystery. Conceivably, quantum mechanics might work, wrote Bell, only when "the settings of the instruments are made sufficiently in advance to allow them to reach some mutual rapport" by exchange of signals at the speed of light. "In that connection, experiments of the type proposed [in 1957] by Bohm and Aharonov, in which the settings are changed during the flight of the particles, are crucial."

The practical problem with this experiment was that the huge, fragile, piles-of-plates polarizers at either end of the Freedman-Clauser experiment could not move into their settings quickly. Aspect had come up with a beautiful (and, importantly, frugal) alternative idea. Its main ingredient was water.

"Each polarizer," Aspect explained to Bell, "would be replaced by a setup involving a switching device followed by two polarizers in two different orientations." At any given time, the switch would allow a path to only one of the two polarizers. "The switch would rapidly redirect the incident light from one polarizer to the other one," leaving no time for light-speed signals to facilitate any kind of "mutual rapport" between distant ends of the apparatus. He turned to the blackboard, where he wrote the appropriate inequality for the situation "if the two switches work at random and are uncorrelated."

Aspect's "switches" were two glass boxes full of water, over forty-two feet apart from each other, on either side of the beam of cascading calcium that produced the photons. Each box of water carried a sound wave, far higher than the human ear can hear. (Transducers on either side of the boxes converted electrical signals into this ultrasonic wave.)

Sound waves, unlike light waves, need a medium—hence the silence of outer space. They operate by repeatedly compressing and then relieving pressure in their medium, so that the air or water they move through becomes alternately denser and thinner, denser and thinner. When the water is thinned by the ultrasonic wave, photons can pass through to the polarizer beyond; when it is dense, it deflects the photons to the other polarizer, set at the alternate setting. The wave cycles far faster than a photon can travel the twenty-one feet separating the source from the switch. "The switching between the two channels would occur about every ten nanoseconds," Aspect explained. Meanwhile, it would take a light signal four times that long to travel between the two locations.

It would not, he admitted, be the ideal scheme, "since the change is not truly random, but rather quasiperiodic. Nevertheless, the two switches on the two sides would be driven by different generators at different frequencies," meaning that the two boxes would oscillate at different rates, and in practice the rates would drift. "It is then very natural," said Aspect, "to assume that they function in an uncorrelated way."*

When Aspect finished his eager presentation, he stood silently awaiting a reply. Bell asked his first question with a trace of irony: "Have you a permanent position?" Aspect was only a graduate student, but—because of the uniqueness of the French system, and in drastic contrast to his counterparts in America—his position at the École normale supérieure was actually permanent. Even with this advantage, it was not easy.

"There will be serious fights," Bell warned him. But the stigma was not the only thing he worried about. "One should not spend all his time on concepts. You are an experimentalist, which keeps your feet on the ground, so you are not in so much danger. For me, I am a theorist and this subject must remain my hobby.

"If you spend all your time thinking about it, you are in danger of becoming crazy."

Freedman drifted away from Bell physics, but he found his thesis experiment haunting him even thirty years later. "The Bell experiment was a null experiment—that's an experiment that measures no deviation from what you expected—and in my time here, I've done twenty-four null experiments, finding that those things that you didn't expect to be there are, in fact, *not* there. So this was sort of how I started my career."

The "constant theme" of Freedman's career was, as he said in 2000, "It's really a big help if you know what the right answer should be: if you don't get it, you might suspect that there's something wrong with your equipment—and that's probably the case.

"I have quite a reputation for doing this, for stepping into a field

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*This experiment (published in *Physical Review Letters* for Christmas 1982) was so difficult to carry out that Aspect and his student, Jean Dalibard, listed the machinist, Gérard Roger, as an author.

where there's something very exciting going on"—Freedman grinned—"and leaving it with nothing interesting." The last word on the subject was far from spoken, however.

Holt, too, left Bell and EPR behind for the "bread-and-butter physics" his apparatus had once been designed to do. He went on to a career of measuring atomic lifetimes via cascade photons, measuring spectra with lasers, and measuring energy levels in atoms too complicated for quantum mechanics (essentially any atom beyond hydrogen). Looking back, "I would say that I played a relatively minor role in the CHSH [Clauser-Horne-Shimony-Holt] business," Holt said. Then he grinned. "But I did set the world on fire with my wrong result."

As it had with Freedman, the experience started him contemplating how science proceeds. "There's an interesting scientific principle that a wrong answer can be much more stimulating to the field than just sort of finding the answer that's in the back of the book. A wrong result gets people excited. Worried.

"Obviously, you don't *really* want that to be happening—it's O.K. for a *theorist* to come up with a speculative new theory that gets shot down, but *experimentalists* are supposed to be very careful and their error limits are supposed to be realistic. Unfortunately, with this experiment, whenever you're looking for a stronger correlation, any kind of systematic error you can imagine typically weakens it and moves it toward the hidden-variable range. It was a hard experiment. In those days, at any rate, with the kind of equipment I had, and . . . well, what can I say?" He laughed with a shrug. "I screwed up."

Clarity, however, about which experiment is right is not the same as clarity about the quantum mechanics these experiments were designed to elucidate. "The thing is," Holt said, "I'm a scientist and I sort of want to believe what Nature says is the answer and not what I just think I know ahead of time, and I've always thought that quantum mechanics was just so wonderful because it's a surprise—it's sort of this hidden knowledge that we high priests of science in a way"—he laughed—"can find. . . .

"Which isn't to say I want to keep it secret," he explained. "But if everything were just obvious, if you could just look around and see how the universe is, well, that wouldn't be very interesting. Quantum mechanics is very subtle; that's its fascination.

"For me, well—*all* of my interest in physics has been because I personally wanted to know the answer to these questions . . . and," he said simply and a little wistfully, "I still don't know the answers. . . . That's the frustrating thing. . . . I think it's going to be a long time before people know what quantum mechanics means. All these modern experiments *clearly* show that you have to at least provisionally accept the quantum-mechanical way of thinking about states of unobserved things. And yet it's a very unsatisfying kind of thing.

"Quantum mechanics—how you talk about it, that is—still remains unfinished business. But I believe something will come up, from an unexpected direction. . . . We may not have to solve the problem as originally posed at all—the problem will just disappear one day because we'll find out we were asking the wrong question."

In 1975, Mike Horne thought he was leaving Bell physics behind, too. He and Shimony had become fascinated by a beautiful new experimental apparatus, a neutron interferometer, just invented by Helmut Rauch in Vienna. In contrast to Clauser's experiment exhibiting the particle nature of light, Rauch's machine dramatically displayed the wave nature of matter.

In the first years of the nineteenth century, as Horne vividly describes it, the great physicist Thomas Young "showed experimentally that the addition [or superposition] of two equally bright light beams can make darkness, and, under slightly different conditions, can make light four times brighter than either original beam." Horne smiles: "That is, 1 plus 1 equals 0, and, under other conditions, equals 4." This is called interference, and it signifies the presence of a wave.

But Rauch was displaying these hallmark wave phenomena with particles of matter. Beams of neutrons, particles produced from the seething hot core of a nuclear reactor, were interfering with each other like waves.

The neutron interferometer offered these neutrons two alternative V-shaped paths, like a child choosing to throw a ball to his friend by bouncing it off the floor or off a low ceiling. Whether, after entering the interferometer, a neutron hit the "floor" or the "ceiling," its final destination was the same, so two neutrons, starting at the same point but taking alternative paths, would meet again afterward. These alternative paths seen together traced out a diamond shape. When they met, the neutrons would interfere with each other.

Dramatically, even a *single* neutron in the interferometer would interfere with itself. This, like so many quantum-mechanical conundrums, is impossible to picture. It is as if one neutron had traveled down both paths simultaneously.

Horne and Shimony had been immersed for a decade in the mysteries of two-particle entanglement; now they were distracted by this magical single-particle effect. "Abner and I both thought, This is going to be a very

The Center for History of Physics of the American Institute of Physics has tape recorded interviews with transcripts that are available on the internet. These provide interesting historical insights and two that are particularly relevant are one with John Clauser and one with Abner Shimony:

“Interview of John Francis Clauser by Joan Bromberg on May 20, 2002,” in *The Niels Bohr Library & Archives*, (American Institute of Physics, College Park, MD USA). http://www.aip.org/history/ohilist/25096.html

“Interview of Abner Shimony by Joan Bromberg on September 9, 2002,” in *The Niels Bohr Library & Archives*, (American Institute of Physics, College Park, MD USA). http://www.aip.org/history/ohilist/25643.html

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Bell's theorem: experimental tests and implications

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Abstract

Bell's theorem represents a significant advance in understanding the conceptual foundations of quantum mechanics. The theorem shows that essentially all local theories of natural phenomena that are formulated within the framework of realism may be tested using a single experimental arrangement. Moreover, the predictions by these theories must significantly differ from those by quantum mechanics. Experimental results evidently refute the theorem's predictions for these theories and favour those of quantum mechanics. The conclusions are philosophically startling: either one must totally abandon the realistic philosophy of most working scientists, or dramatically revise our concept of space–time.

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1. Introduction

Realism is a philosophical view, according to which external reality is assumed to exist and have definite properties, whether or not they are observed by someone. So entrenched is this viewpoint in modern thinking that many scientists and philosophers have sought to devise conceptual foundations for quantum mechanics that are clearly consistent with it. One possibility, it has been hoped, is to reinterpret quantum mechanics in terms of a statistical account of an underlying hidden-variables theory in order to bring it within the general framework of classical physics. However, Bell's theorem has recently shown that this cannot be done. The theorem proves that all realistic theories, satisfying a very simple and natural condition called locality, may be tested with a single experiment against quantum mechanics. These two alternatives necessarily lead to significantly different predictions. The theorem has thus inspired various experiments, most of which have yielded results in excellent agreement with quantum mechanics, but in disagreement with the family of local realistic theories. Consequently, it can now be asserted with reasonable confidence that either the thesis of realism or that of locality must be abandoned. Either choice will drastically change our concepts of reality and of space–time.

The historical background for this result is interesting, and represents an extreme irony for Einstein's steadfastly realistic position, coupled with his desire that physics be expressable solely in simple geometric terms. Within the realistic framework, Einstein *et al* (1935, hereafter referred to as EPR) presented a classic argument. As a starting point, they assumed the non-existence of action-at-a-distance and that some of the statistical predictions of quantum mechanics are correct. They considered a system consisting of two spatially separated but quantum-mechanically correlated particles. For this system, they showed that the results of various experiments are predetermined, but that this fact is not part of the quantum-mechanical description of the associated systems. Hence that description is an incomplete one. To complete the description, it is thus necessary to postulate additional 'hidden variables', which presumably will then restore completeness, determinism and causality to the theory.

Many in the physics community rejected their argument, preferring to follow a counter-argument by Bohr (1935), who believed that the whole realistic viewpoint is inapplicable. Many others, however, felt that since both viewpoints lead to the same observable phenomenology, a commitment to either one is only a matter of taste. Hence, the discussion, for the greater part of the subsequent 30 years, was pursued perhaps more at physicists' cocktail parties than in the mainstream of modern research.

Starting in 1965, however, the situation changed dramatically. Using essentially the same postulates as those of EPR, J S Bell showed for a *Gedankenexperiment* of Bohm (a variant of that of EPR) that no deterministic local hidden-variables theory can reproduce all of the statistical predictions by quantum mechanics. Inspired by that work, Clauser *et al* (1969, hereafter referred to as CHSH) added three contributions. First, they showed that his analysis can be extended to cover actual systems, and that experimental tests of this broad class of theories can be performed. Second, they introduced a very reasonable auxiliary assumption which allows tests to be performed with existing technology. Third, they specifically proposed performing such a test by examining the polarisations of photons produced by an atomic cascade, and derived the required conditions for such an experiment.

Curiously, the transition to a consideration of real systems introduced new aspects to the problem. EPR had demonstrated that any ideal system which satisfies a locality condition must be deterministic (at least with respect to the correlated properties). Since that argument applies only to ideal systems, CHSH therefore had postulated determinism explicitly. Yet, it eventually became clear that it is not the deterministic character of these theories that is incompatible with quantum mechanics. Although not stressed, this point was contained in Bell's subsequent papers (1971, 1972)—any non-deterministic (stochastic) theory satisfying a more general locality condition is also incompatible with quantum mechanics. Indeed it is the objectivity of the associated systems and their locality which produces the incompatibility. Thus, the whole realistic philosophy is in question! Bell's (1971) result, however, is in a form that is awkward for an experimental test. To facilitate such tests, Clauser and Horne (1974, hereafter referred to as CH) explicitly characterised this broad class of theories. They then gave a new incompatibility theorem that yields an experimentally testable result and derived the requirements for such a test. Although such an experiment is difficult to perform (and in fact has not yet been performed), they showed that an assumption weaker in certain respects than the one of CHSH allowed the experiments proposed earlier by CHSH to be used as a test for these theories also.

The interpretation of all of the existing results requires at least some auxiliary assumptions, although experiments are possible for which this is not the case. Even though some of the assumptions are very reasonable, this fact allows loopholes still to exist. Experiments now in progress or being planned will be able to eliminate most of these loopholes. However, even now one can assert with reasonable confidence that the experimental evidence to date is contrary to the family of local realistic theories. The construction of a quantum-mechanical world view as an alternative to the point of view of the local realistic theories is beyond the scope of this review.

Section 2 of this review summarises the argument of EPR, appendix 1 discusses various critical evaluations of it, and appendix 2 summarises briefly the history of hidden-variables theories. Section 3 describes the versions of Bell's theorem discussed above as well as some others. Section 4 discusses the requirements for a fully general test and shows why such an experiment is a difficult one to perform. Section 5 is devoted to a description of the cascade-photon experiments proposed by CHSH. First, it discusses the auxiliary assumptions by CHSH and CH. Second, calculations of the quantum-mechanical predictions for these experiments are summarised. Third, there is a discussion of the actual cascade-photon experiments performed so far (Freedman and Clauser 1972, Holt and Pipkin 1973, Clauser 1976, Fry and Thompson 1976). All but the second agree very well with the quantum-mechanical predictions, thus providing significant evidence against the entire family of local realistic theories. Section 5 ends with a critique of the CH and CHSH assumptions. Section 6 summarises and discusses related experiments measuring the polarisation correlation of photons produced in positronium annihilation (Kasday *et al* 1975, Faraci *et al* 1974, Wilson *et al* 1976, Bruno *et al* 1977) and an experiment measuring the spin correlation of proton pairs (Lamehi-Rachti and Mittig 1976). Section 7 is devoted to an evaluation of the experimental results obtained so far and to the prospects for future experiments.

5.3.1. Experiment by Freedman and Clauser (1972). Freedman and Clauser (1972, see also Freedman 1972) observed the 5513 Å and 4227 Å pairs produced by the $4p^2\ {}^1S_0 \rightarrow 4p4s\ {}^1P_1 \rightarrow 4s^2\ {}^1S_0$ cascade in calcium. Their arrangement is shown schematically in figure 5. Calcium atoms in a beam from an oven were excited by resonance absorption to the $3d4p\ {}^1P_1$ level, from which a considerable fraction decayed to the $4p^2\ {}^1S_0$ state at the top of the cascade. No precaution was necessary for eliminating isotopes with non-zero nuclear spin, since 99·855% of naturally occurring calcium has zero nuclear spin. Pile-of-plates polarisation analysers were used, with transmittances $\epsilon_M{}^1 = 0{\cdot}97 \pm 0{\cdot}01$, $\epsilon_m{}^1 = 0{\cdot}038 \pm 0{\cdot}004$, $\epsilon_M{}^2 = 0{\cdot}96 \pm 0{\cdot}01$, $\epsilon_m{}^2 = 0{\cdot}037 \pm 0{\cdot}004$. Each analyser could be rotated by angular increments of $\pi/8$, and the plates could be folded out of the optical path on hinged frames. The half-angle ξ subtended by the primary lenses was 30°. Coincidence counting was done for 100 s periods; periods during which all plates were removed alternated with periods during which all were inserted. In each run the ratios $R(\pi/8)/R_0$ and $R(3\pi/8)/R_0$ were determined. Corrections were made for accidental coincidences, but even without this correction, the results still significantly violated inequality (5.6). The average ratios for roughly 200 h of running time are:

$$[R(\pi/8)/R_0]_{\text{expt}} = 0{\cdot}400 \pm 0{\cdot}007 \qquad [R(3\pi/8)/R_0]_{\text{expt}} = 0{\cdot}100 \pm 0{\cdot}003$$

and therefore:

$$[R(\pi/8)/R_0 - R(3\pi/8)/R_0]_{\text{expt}} = 0{\cdot}300 \pm 0{\cdot}008$$

in clear disagreement with inequality (5.6). The quantum-mechanical predictions are obtained from equation (5.15) (with allowances for uncertainties in the measurement of the transmittances and the subtended angle):

$$[R(\pi/8)/R_0 - R(3\pi/8)/R_0]_{\text{QM}} = (0{\cdot}401 \pm 0{\cdot}005) - (0{\cdot}100 \pm 0{\cdot}005) = 0{\cdot}301 \pm 0{\cdot}007.$$

The agreement between the experimental results with the quantum-mechanical predictions is excellent. Agreement is also found for other values of the angle ϕ, as well as for measurements made with only one or the other polariser removed.

5.3.2. Experiment by Holt and Pipkin (1973). Holt and Pipkin (1973, see also Holt 1973) observed 5676 Å and 4047 Å photon pairs produced by the $9^1P_1 \rightarrow 7^3S_1 \rightarrow 6^3P_0$ cascade in the zero nuclear-spin isotope ^{198}Hg (see figure 6 for a partial level diagram of mercury). Atoms were excited to the 9^1P_1 level by a 100 eV electron beam. The density matrix of the 9^1P_1 level was found to be approximately $\frac{1}{3} I$ by measurements of the polarisation of the 5676 Å photons, so that equation (5.15) with $F_1(\xi)$ replaced by $-F_2(\xi)$ is used to calculate the quantum-mechanical predictions for the coincidence counting rates. Calcite prisms were employed as polarisation analysers, with measured transmittances:

$$\epsilon_M{}^1 = 0{\cdot}910 \pm 0{\cdot}001 \qquad \epsilon_M{}^2 = 0{\cdot}880 \pm 0{\cdot}001$$

$$\epsilon_m{}^1 < 10^{-4} \qquad \epsilon_m{}^2 < 10^{-4}.$$

The half-angle ξ was taken to be 13° ($F_2(13°) = 0{\cdot}9509$). The quantum-mechanical prediction is:

$$[R(3\pi/8)/R_0 - R(\pi/8)/R_0]_{\text{QM}} = 0{\cdot}333 - 0{\cdot}067 = 0{\cdot}266$$

which only marginally exceeds the value $\frac{1}{4}$ allowed by inequality (5.6). The experimental result in 154·5 h of coincidence counting, however, is:

$$[R(3\pi/8)/R_0 - R(\pi/8)/R_0]_{\text{expt}} = 0{\cdot}316 \pm 0{\cdot}011 - 0{\cdot}099 \pm 0{\cdot}009 = 0{\cdot}216 \pm 0{\cdot}013$$

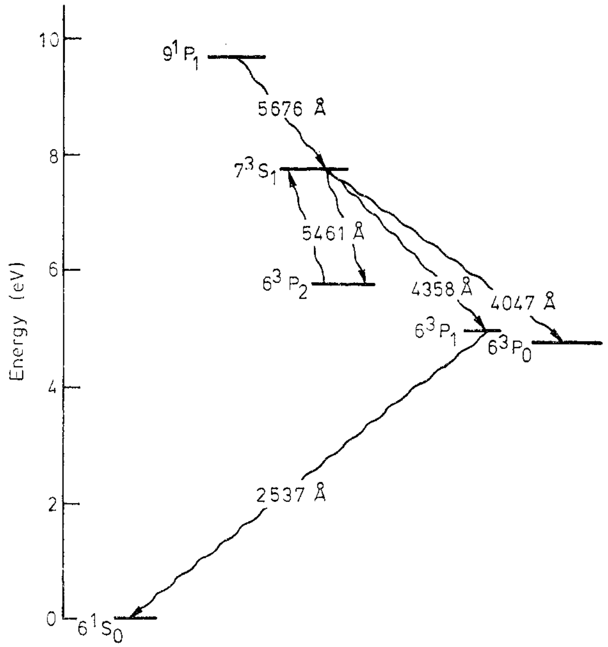

Figure 6. Partial level scheme for atomic mercury. Experiments by Holt and Pipkin, and by Clauser, excited the atoms to the 9^1P_1 level by electron bombardment, and observed photons emitted by the $9^1P_1 \rightarrow 7^3S_1 \rightarrow 6^3P_0$ cascade. The experiment by Fry and Thompson excited atoms to the 6^3P_2 (metastable) level by electron bombardment. Downstream, the atoms were excited by a tunable dye laser to the 7^3S_1 level, and photons were observed from the $7^3S_1 \rightarrow 6^3P_1 \rightarrow 6^1S_0$ cascade.

in good agreement with inequality (5.6) but in sharp disagreement with the quantum-mechanical prediction. Since this result is very surprising, Holt and Pipkin took great care to check possible sources of systematic error: the contamination of the source by isotopes with non-zero nuclear spin, perturbation by external magnetic or electric fields, coherent multiple scattering of the photons (radiation trapping), polarisation sensitivity of the photomultipliers, and spurious counts from residual radioactivity and/or cosmic rays, etc.

One such systematic error was found in the form of stresses in the walls of the Pyrex bulb used to contain the electron gun and mercury vapour. Estimates of the optical activity of these walls were then made, and the results were corrected correspondingly. (The values presented above include this correction.) It is noteworthy, however, that only the retardation sum for both windows was measured, for light entering the cell from one side and exiting through the opposite side. On the other hand, in the present experiment in which light exits from both windows, the relevant quantity is the retardation difference.

It is also noteworthy that in the subsequent experiment by Clauser (§5.3.3), a correlation was first measured which agreed with the results of Holt and Pipkin. Stresses were then found in one lens which were due to an improper mounting. (These were too feeble to be detected by a simple visual check using crossed Polaroids.) The stresses were removed, the experiment was re-performed, and excellent agreement with quantum mechanics was then obtained. On the other hand, Holt and Pipkin did not repeat their experiment when they discovered the stresses in their bulb.

A second criticism is that Holt and Pipkin took the solid-angle limit to be that imposed by a field stop placed outside the collimating lenses. It is possible that lens aberrations may have allowed a larger solid angle than they recognised. A ray-tracing calculation was in fact performed to assure that this was not the case. However, a solid stop ahead of the lens would have given one greater confidence that this did not, in fact, occur.

5.3.3. Experiment by Clauser (1976). Clauser (1976) repeated the experiment of Holt and Pipkin, using the same cascade and same excitation mechanism, though with a source consisting mainly of the zero-spin isotope ^{202}Hg. (The depolarisation effect due to some residual non-zero nuclear spin isotopes was calculated, using some results of Fry (1973).) Pile-of-plates polarisers were used with transmittances:

$$\epsilon_M{}^1 = 0{\cdot}965 \qquad \epsilon_m{}^1 = 0{\cdot}011 \qquad \epsilon_M{}^2 = 0{\cdot}972 \qquad \epsilon_m{}^2 = 0{\cdot}008$$

and the half-angle ξ taken to be 18·6°. The quantum-mechanical prediction is:

$$[R(3\pi/8)/R_0 - R(\pi/8)/R_0]_{QM} = 0{\cdot}2841.$$

The experimental result, from 412 h of integration, is:

$$[R(3\pi/8)/R_0 - R(\pi/8)/R_0]_{\text{expt}} = 0{\cdot}2885 \pm 0{\cdot}0093$$

in excellent agreement with the quantum-mechanical prediction, but in sharp disagreement with inequality (5.6).

5.3.4. Experiment by Fry and Thompson (1976). Fry and Thompson (1976) observed the 4358 Å and 2537 Å photon pairs emitted by the $7^3S_1 \rightarrow 6^3P_1 \rightarrow 6^1S_0$ cascade in the zero nuclear-spin isotope ^{200}Hg. Their experiment is shown schematically in figure 7.

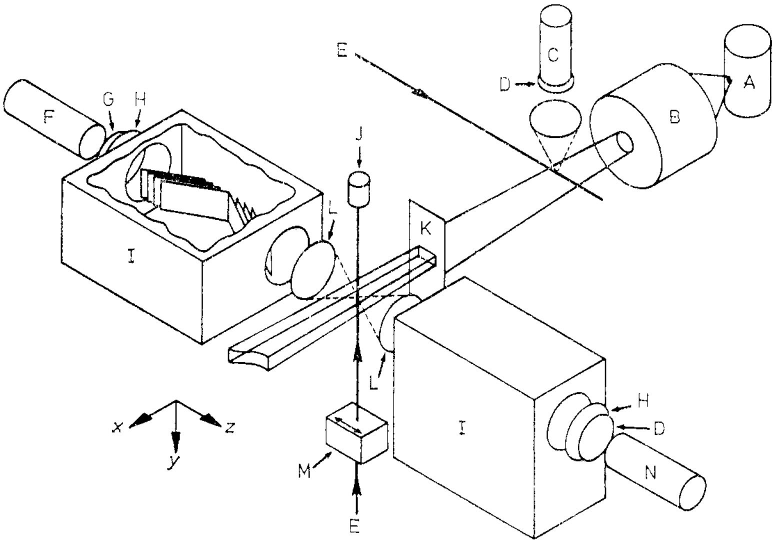

Figure 7. Schematic diagram of the experimental arrangement of Fry and Thompson. Polariser plate arrangement is also indicated. Actual polarisers have 14 plates. A, Hg oven; B, solenoid electron gun; C, RCA 8575; D, 4358 Å filter; E, 5461 Å laser beam; F, Amperex 56 DUVP/03; G, 2537 Å filter; H, focusing lens; I, pile-of-plates polariser; J, laser beam trap; K, atomic beam defining slit; L, light collecting lens; M, crystal polariser; N, RCA 8850 (figure after Fry and Thompson).

An atomic beam consisting of natural mercury was used as a source of ground-state (6^1S_0) atoms. The excitation of these to the 7^3S_1 level occurred in two steps at different locations along the beam. First, the atoms were excited by electron bombardment to the metastable 6^3P_2 level. Downstream, where all rapidly decaying states had vanished, a single isotope was excited to the 7^3S_1 level by resonant absorption of 5461 Å radiation from a narrow-bandwidth tunable dye laser. The technique provided a high data accumulation rate, since only the cascade of interest was excited. Photons were collected over a half-angle ξ of $19{\cdot}9° \pm 0{\cdot}3°$, and pile-of-plates analysers were used, with transmittances:

$$\epsilon_M{}^1 = 0{\cdot}98 \pm 0{\cdot}01 \qquad \epsilon_m{}^1 = 0{\cdot}02 \pm 0{\cdot}005 \qquad \epsilon_M{}^2 = 0{\cdot}97 \pm 0{\cdot}01 \qquad \epsilon_m{}^2 = 0{\cdot}02 \pm 0{\cdot}005.$$

The density matrix of the 7^3S_1 level was ascertained by polarisation measurements of the 4358 Å photons; it was found to be diagonal even though the Zeeman sublevels were not equally populated. The quantum-mechanical prediction is:

$$[R(3\pi/8)/R_0 - R(\pi/8)/R_0]_{\mathrm{QM}} = 0{\cdot}294 \pm 0{\cdot}007.$$

The experimental result is:

$$[R(3\pi/8)/R_0 - R(\pi/8)/R_0]_{\mathrm{expt}} = 0{\cdot}296 \pm 0{\cdot}014$$

in excellent agreement with the quantum-mechanical prediction, but again in sharp disagreement with inequality (5.5). Because of the high pumping rate attainable with the dye laser, it was possible to gather the data in a remarkably short period of 80 min which, of course, diminished the probability of errors due to variations in the operation of the apparatus, and facilitated checking for systematic errors.

Experimental Realization of Einstein-Podolsky-Rosen-Bohm *Gedankenexperiment*: A New Violation of Bell's Inequalities

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The linear-polarization correlation of pairs of photons emitted in a radiative cascade of calcium has been measured. The new experimental scheme, using two-channel polarizers (i.e., optical analogs of Stern-Gerlach filters), is a straightforward transposition of Einstein-Podolsky-Rosen-Bohm *gedankenexperiment*. The present results, in excellent agreement with the quantum mechanical predictions, lead to the greatest violation of generalized Bell's inequalities ever achieved.

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In the well-known Einstein-Podolsky-Rosen-Bohm *gedankenexperiment*[1] (Fig. 1), a source emits pairs of spin-$\frac{1}{2}$ particles, in a singlet state (or pairs of photons in a similar nonfactorizing state). After the particles have separated, one performs correlated measurements of their spin components along arbitrary directions $\vec{a}$ and $\vec{b}$. Each measurement can yield two results, denoted ±1; for photons, a measurement along $\vec{a}$ yields the result +1 if the polarization is found parallel to $\vec{a}$, and −1 if the polarization is found perpendicular. For a singlet state, quantum mechanics predicts some correlation between such measurements on the two particles. Let us denote by $P_{\pm\pm}(\vec{a},\vec{b})$ the probabilities of obtaining the result ±1 along $\vec{a}$ (particle 1) and ±1 along $\vec{b}$ (particle 2). The quantity

$$E(\vec{a},\vec{b})=P_{++}(\vec{a},\vec{b})+P_{--}(\vec{a},\vec{b})-P_{+-}(\vec{a},\vec{b})-P_{-+}(\vec{a},\vec{b}) \tag{1}$$

is the correlation coefficient of the measurements on the two particles. Bell[2] considered theories explaining such correlations as due to common properties of both particles of the same pair; adding a locality assumption, he showed that they are constrained by certain inequalities that are not always obeyed by the predictions of quantum mechanics. Such theories are called[3] "realistic local theories" and they lead to the generalized Bell's inequalities[4]

$$-2 \leqslant S \leqslant 2, \tag{2}$$

where

$$S=E(\vec{a},\vec{b})-E(\vec{a},\vec{b}\,')+E(\vec{a}\,',\vec{b})+E(\vec{a}\,',\vec{b}\,')$$

involves four measurements in four various orientations. On the other hand, for suitable sets of orientations,[4] the quantum mechanical predictions can reach the values $S=\pm2\sqrt{2}$, in clear contradiction with (2): Quantum mechanics cannot be completed by an underlying structure such as "realistic local theories."

Several experiments with increasing accuracy have been performed, and they clearly favor quantum mechanics.[3,5] Unfortunately, none allowed a direct test using inequalities (2), since none followed the scheme of Fig. 1 closely enough. Some experiments were performed with pairs of photons (or of protons). But no efficient analyzers are available at such energies, and the results that would have been obtained with ideal polarizers are deduced indirectly from Compton scattering experiments. The validity of such a procedure in the context of Bell's theorem has been criticized.[3,6]

There are also experiments with pairs of low-energy photons emitted in atomic radiative cascades. True polarizers are available in the visible range. However, all previous experiments involved single-channel analyzers, transmitting one polarization ($\vec{a}$ or $\vec{b}$) and blocking the orthog-

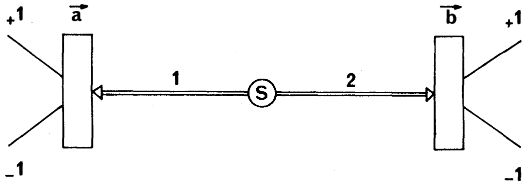

FIG. 1. Einstein-Podolsky-Rosen-Bohm *gedankenexperiment.* Two-spin-$\frac{1}{2}$ particles (or photons) in a singlet state (or similar) separate. The spin components (or linear polarizations) of 1 and 2 are measured along $\vec{a}$ and $\vec{b}$. Quantum mechanics predicts strong correlations between these measurements.

onal one. The measured quantities were thus only the coincidence rates in +1 channels: $R_{++}(\vec{a},\vec{b})$. Several difficulties then arise[3] as a result of the very low efficiency of the detection system (the photomultipliers have low quantum efficiencies and the angular acceptance is small). The measurements of polarization are inherently incomplete: When a pair has been emitted, if no count is obtained at one of the photomultipliers, there is no way to know whether it has been missed by the (low-efficiency) detector or whether it has been blocked by the polarizer (only the latter case would be a real polarization measurement). Thus, coincidence counting rates such as $R_{+-}(\vec{a},\vec{b})$ or $R_{--}(\vec{a},\vec{b})$ cannot be measured directly. It is nevertheless possible to derive from the experimental data numerical quantities which can (according to quantum mechanics) possibly violate Bell-type inequalities. For this purpose, one has to resort to auxiliary experiments, where coincidence rates are measured with one or both polarizers removed. Some reasoning, with a few additional—and very natural—assumptions (such as the "no-enhancement" assumption of Clauser and Horne[7]), then allows one to obtain actually operational inequalities.

In this Letter, we report the results of an experiment following much more closely the ideal scheme of Fig. 1. True dichotomic polarization measurements on visible photons have been performed by replacing ordinary polarizers by two-channel polarizers, separating two orthogonal linear polarizations, followed by two photomultipliers (Fig. 2). The polarization measurements then become very similar to usual Stern-Gerlach measurements for spin-$\frac{1}{2}$ particles.[8]

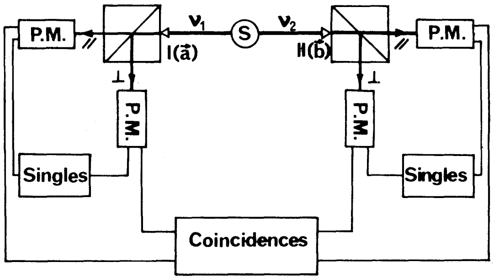

FIG. 2. Experimental setup. Two polarimeters I and II, in orientations $\vec{a}$ and $\vec{b}$, perform true dichotomic measurements of linear polarization on photons ν_1 and ν_2. Each polarimeter is rotatable around the axis of the incident beam. The counting electronics monitors the singles and the coincidences.

Using a fourfold coincidence technique, we measure in a single run the four coincidence rates $R_{\pm\pm}(\vec{a},\vec{b})$, yielding directly the correlation coefficient for the measurements along $\vec{a}$ and $\vec{b}$:

$$E(\vec{a},\vec{b})=\frac{R_{++}(\vec{a},\vec{b})+R_{--}(\vec{a},\vec{b})-R_{+-}(\vec{a},\vec{b})-R_{-+}(\vec{a},\vec{b})}{R_{++}(a,b)+R_{--}(a,b)+R_{+-}(a,b)+R_{-+}(a,b)}. \qquad (3)$$

It is then sufficient to repeat the same measurements for three other choices of orientations, and inequalities (2) can directly be used as a test of realistic local theories versus quantum mechanics. This procedure is sound if the measured values (3) of the correlation coefficients can be taken equal to the definition (1), i.e., if we assume that the ensemble of actually detected pairs is a faithful sample of all emitted pairs. This assumption is highly reasonable with our very symmetrical scheme, where the two measurement results +1 and −1 are treated in the same way (the detection effiencies in both channels of a polarizer are equal). All data are collected in very similar experimental conditions, the only changes being rotations of the polarizers.

Such a procedure allows us not only to suppress possible systematic errors (e.g., changes occurring when removing the polarizers) but also to control more experimental parameters. For instance, we have checked that the sum of the coincidence rates of one photomultiplier with both photomultipliers on the other side is constant. We have also observed that the sum of the four coincidence rates $R_{\pm\pm}(\vec{a},\vec{b})$ is constant when changing the orientations; thus the size of the selected sample is found constant.

We have used the high-efficiency source previously described.[5] A $(J=0)\rightarrow(J=1)\rightarrow(J=0)$ cascade in calcium-40 is selectively excited by two-photon absorption, with use of two single-mode lasers. Pairs of photons (at wavelengths $\lambda_1=551.3$ nm and $\lambda_2=422.7$ nm) correlated in polarization are emitted at a typical rate of 5×10^7 s^{-1}. The polarizers are polarizing cubes (Fig. 2) made of two prisms with suitable dielectric thin films on the sides stuck together; the faces are antirefelction coated. Cube I transmits light polarized in the incidence plane onto the active hypotenuse (parallel polarization, along $\vec{a}$) while it reflects the orthogonal polarization (perpendicular polarization). Cube II works similarly. For actual polarizers we define transmission and reflection coefficients: $T^{\parallel}$ and $R^{\perp}$ are close to 1, while $T^{\perp}$ and $R^{\parallel}$ are close to 0. The measured values of our devices are $T_1^{\parallel}=R_1^{\perp}=0.950$ and $T_1^{\perp}=R_1^{\parallel}=0.007$ at λ_1; $T_2^{\parallel}=R_2^{\perp}=0.930$ and $T_2^{\perp}=R_2^{\parallel}=0.007$ at λ_2 (all values are ± 0.005). Each polarizer is mounted in a rotatable mechanism holding two photomultipliers; we call the ensemble a polarimeter. The gains of the two photomultipliers are adjusted for the equality of the counting detection efficiencies in both channels of a polarimeter (2×10^{-3} at 422 nm, 10^{-3} at 551 nm). Typical single rates (over 10^4 s^{-1}) are high compared with dark rates (10^2 s^{-1}). Wavelength filters at 422 or 551 nm are mounted in front of each photomultiplier. The fourfold coincidence electronics includes four overlap-type coincidence circuits. Each coincidence window, about 20 ns wide, has been accurately measured. Since they are large compared to the lifetime of the intermediate state of the cascade (5 ns) all true coincidences are registered. We infer the accidental coincidence rates from the corresponding single rates, knowing the widths of the windows. This method is valid with our very stable source, and it has been checked by comparing it with the methods of Ref. 5, using delayed coincidence channels and/or a time-to-amplitude converter. By subtraction of these accidental rates (about 10 s^{-1}) from the total rates, we obtain the true coincidence rates $R_{\pm\pm}(\vec{a},\vec{b})$ (actual values are in the range 0–40 s^{-1}, depending on the orientations). A run lasts 100 s, and $E(\vec{a},\vec{b})$ derived from Eq. (3) is measured with a typical statistical accuracy of ±0.02 (the sum of the four coincidence rates is typically 80 s^{-1}).

It is well known that the greatest conflict between quantum mechanical predictions and the inequalities (2) is expected for the set of orientations $(\vec{a},\vec{b})=(\vec{b},\vec{a}\,')=(\vec{a}\,',\vec{b}\,')=22.5°$ and $(\vec{a},\vec{b}\,')=67.5°$. Five runs have been performed at each of these orientations; the average yields

$$S_{\mathrm{expt}}=2.697\pm0.015. \tag{4}$$

The indicated uncertainty is the standard deviation accounting for the Poisson law in photon counting. The impressive violation of inequalities (2) is 83% of the maximum violation predicted by quantum mechanics with ideal polarizers (the largest violation of generalized Bell's inequalities previously reported was 55% of the predicted violation in the ideal case[5]).

With symmetrical polarimeters, quantum mechanics predicts

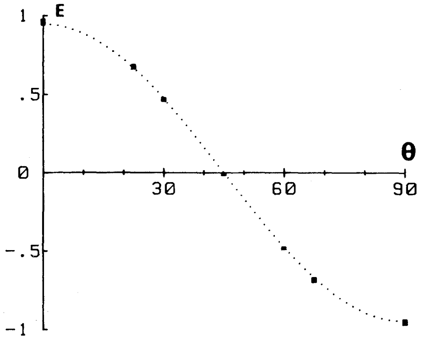

FIG. 3. Correlation of polarizations as a function of the relative angle of the polarimeters. The indicated errors are ±2 standard deviations. The dotted curve is not a fit to the data, but quantum mechanical predictions for the actual experiment. For ideal polarizers, the curve would reach the values ±1.

$$E(\vec{a},\vec{b})=F\,\frac{(T_1^{\parallel}-T_1^{\perp})(T_2^{\parallel}-T_2^{\perp})}{(T_1^{\parallel}+T_1^{\perp})(T_2^{\parallel}+T_2^{\perp})}\cos 2(\vec{a},\vec{b}). \tag{5}$$

($F=0.984$ in our case; it accounts for the finite solid angles of detection.) Thus, for our experiment,

$$S_{\mathrm{QM}}=2.70\pm0.05. \tag{6}$$

The indicated uncertainty accounts for a slight lack of symmetry between both channels of a polarimeter: We have found a variation of ±1% of the detection efficiencies when rotating the polarimeters. This spurious effect has been explained as small displacements of the light beam impinging onto the photocathode. The effect of these variations on the quantum mechanical predictions has been computed, and cannot create a variation of S_{QM} greater than 2%.[9]

Figure 3 shows a comparison of our results with the predictions of quantum mechanics. Here, for each relative orientation $\theta=(\vec{a},\vec{b})$, we have averaged several measurements in different absolute orientations of the polarimeters; this procedure averages out the effect of the slight variations of the detection efficiencies with orientation. The agreement with quantum mechanics is better than 1%.

In conclusion, our experiment yields the strongest violation of Bell's inequalities ever achieved, and excellent agreement with quantum mechanics. Since it is a straightforward transposition of the ideal Einstein-Podolsky-Rosen-Bohm scheme, the experimental procedure is very simple, and needs no auxiliary measurements as in previous experiments with single-channel polarizers. We are thus led to the rejection of realistic local theories if we accept the assumption that there is no bias in the detected samples: Experiments support this natural assumption.

Only two loopholes remain open for advocates of realistic theories without action at a distance.[10] The first one, exploiting the low efficiencies of detectors, could be ruled out by a feasible experiment.[11] The second one, exploiting the static character of all previous experiments, could also be ruled out by a "timing experiment" with variable analyzers[12] now in progress.

The authors acknowledge many valuable discussions with F. Laloë about the principle of this experiment. They are grateful to C. Imbert who sponsors this work.

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Bell's inequality test: more ideal than ever

Alain Aspect

The experimental violation of Bell's inequalities confirms that a pair of entangled photons separated by hundreds of metres must be considered a single non-separable object — it is impossible to assign local physical reality to each photon.

Bell's theorem[1], formulated in 1964, is one of the profound scientific discoveries of the century. Based on the Einstein, Podolsky and Rosen (EPR) *gedanken*, or thought, experiment[2], it shifted the arguments about the physical reality of quantum systems from the realm of philosophy to the domain of experimental physics. For almost three decades, experimental tests[3] of Bell's inequalities have evolved closer and closer to the ideal EPR scheme. An experiment at the University of Innsbruck[4] has, for the first time, fully enforced Bell's requirement for strict relativistic separation between measurements.

It all started when Einstein *et al.* pointed out that for certain quantum states (described almost simultaneously by Schrödinger, who coined the expression 'quantum entanglement'), quantum mechanics predicts a strong correlation between distant measurements. Figure 1 shows a modern version of the EPR situation, where a pair of entangled photons ν_1 and ν_2 are travelling in opposite directions away from a source. Results of polarization measurements with both polarizers aligned are 100% correlated. That is, each photon may be found randomly either in channel + or − of the corresponding polarizer, but when photon ν_1 is found positively polarized, then its twin companion ν_2 is also found positively polarized. Because no signal can connect the two measurements if it travels at a velocity less than or equal to the speed of light, *c*, and because the choice of the direction of analysis can be made at the very last moment before measurement while the photons are in flight, how — argued Einstein — could one avoid the conclusion that each photon is carrying a property, determining the polarization outcome for any direction of analysis?

This seemingly logical conclusion provides a simple image to understand the correlations between distant and simultaneous measurements. But it means specifying supplementary properties ('elements of reality' in the words of Einstein) beyond the quantum-mechanical description. To the question "Can a quantum-mechanical description of physical reality be considered complete?"[2] Einstein's answer was clearly negative, but this conclusion was incompatible with the 'Copenhagen interpretation' defended by Bohr, for whom the quantum-mechanical description was the ultimate one[5]. This debate between Einstein and Bohr lasted until the end of their lives. As it was, it could hardly be settled, because there was no apparent disagreement on the correlations predicted for an EPR *gedanken* experiment. The point under discussion was the world-view implied by the analysis of the situation.

Bell's theorem changed the nature of the debate. In a simple and illuminating paper[1], Bell proved that Einstein's point of view (local realism) leads to algebraic predictions (the celebrated Bell's inequality) that are contradicted by the quantum-mechanical predictions for an EPR *gedanken* experiment involving several polarizer orientations. The issue was no longer a matter of taste, or epistemological position: it was a quantitative question that could be answered experimentally, at least in principle.

Prompted by the Clauser–Horne–Shimony–Holt paper[6] that framed Bell's inequalities in a way better suited to real experiments, a first series of tests[7], using photon pairs produced in atomic radiative cascades, was performed in the early 1970s at Berkeley, Harvard and Texas A&M. Most results agreed with quantum mechanics, but the schemes used were far from ideal; in particular, the use of single-channel polarizers only gave access to the + outcome. Progress in laser physics and modern optics led to a new generation of experiments carried out by colleagues and myself at Orsay in the early 1980s. They were based on a highly efficient source of pairs of correlated photons, produced by non-linear laser excitations of an atomic radiative cascade. An experiment involving two-channel polarizers, as in the ideal EPR *gedanken* experiment, gave an unambiguous violation of Bell's inequalities by tens of standard deviations, and an impressive agreement with quantum mechanics[8].

A third generation of tests, begun in the late 1980s at Maryland and Rochester[9,10], used nonlinear splitting of ultraviolet photons to produce pairs of correlated EPR photons. With such pairs, measurements can bear either on discrete variables such as polarization or spin components, as considered by Bell, or on continuous variables of the type originally considered by Einstein, Podolsky and Rosen, and studied at Caltech[11]. A remarkable feature of such photon sources is the production of two narrow beams of correlated photons that can be fed into two optical fibres, allowing for tests with great distances between the source and the measuring apparatus, as demonstrated over four kilometres in Malvern[12] and over tens of kilometres in Geneva[13].

The experimenters at Innsbruck[4] used this method to address a fundamental point raised by Bell. In the experiment shown in Fig. 1, where the polarizers' orientations are kept fixed during a run, it is possible to rec-

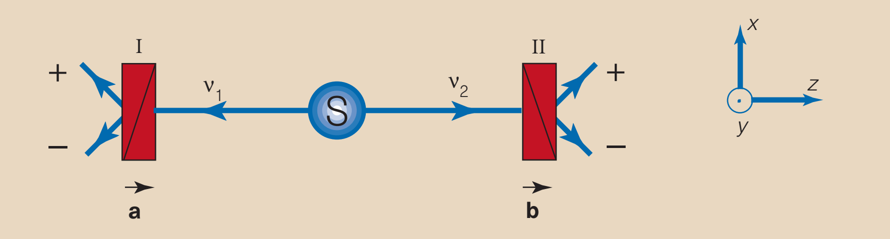

Figure 1 **Einstein–Podolsky–Rosen *gedanken* experiment with photons. The two photons, ν_1 and ν_2, are analysed by the linear polarizers I and II, which make polarization measurements along $\vec{a}$ and $\vec{b}$ perpendicular to the z axis. Each measurement has two possible outcomes, + or −, and one can measure the probabilities of single or joint measurements at various orientations $\vec{a}$ and $\vec{b}$. For an entangled EPR state, violation of a Bell's inequality indicates that the strong correlations between the measurements on the two opposite sides cannot be explained by an image '*à la* Einstein' involving properties carried along by each photon. In the Innsbruck experiment[4], any possibility of communication between the polarizers, at a velocity less than or equal to that of light, is precluded by random and ultrafast switching of the orientations of the polarizers, separated by a distance of 400 m. On each side, a local computer registers the polarizer orientation and the result of each measurement, with the timing monitored by an atomic clock. Data are gathered and compared for correlation measurements after the end of a run.**

oncile the quantum mechanical predictions and Einstein's conceptions by invoking a possible exchange of signals between the polarizers. To avoid this loophole, Bell stressed the importance of experiments "in which the settings are changed during the flight of the particles"[1], so that any direct signal exchange between polarizers would be impossible, provided that the choice of orientations is made randomly in a time shorter than the flight time of the particle or photon, to ensure that relativistic separation is enforced.

Prompted by Bell's remark, a first step towards the realization of this ideal scheme[14] found a violation of Bell's inequality with rapidly switched polarizers, but the polarizer separation (12 m) was too small to allow for a truly random resetting of the polarizers. With a separation of 400 m between their measuring stations, the physicists of Innsbruck[4] have 1.3 μs to make random settings of the polarizer and to register the result of the measurement, as well as its exact timing monitored by a local rubidium atomic clock. It is only at the end of the run that the experimentalists gather the two series of data obtained on each side, and look for correlations. The results, in excellent agreement with the quantum mechanical predictions, show an unquestionable violation of Bell's inequalities[4].

This experiment is remarkably close to the ideal *gedanken* experiment, used to discuss the implications of Bell's theorem. Note that there remains another loophole, due to the limited efficiency of the detectors, but this can be closed by a technological advance that seems plausible in the foreseeable future, and so does not correspond to a radical change in the scheme of the experiment. Although such an experiment is highly desirable, we can assume for the sake of argument that the present results will remain unchanged with high-efficiency detectors.

The violation of Bell's inequality, with strict relativistic separation between the chosen measurements, means that it is impossible to maintain the image '*à la* Einstein' where correlations are explained by common properties determined at the common source and subsequently carried along by each photon. We must conclude that an entangled EPR photon pair is a non-separable object; that is, it is impossible to assign individual local properties (local physical reality) to each photon. In some sense, both photons keep in contact through space and time.

It is worth emphasizing that non-separability, which is at the roots of quantum teleportation[15], does not imply the possibility of practical faster-than-light communication. An observer sitting behind a polarizer only sees an apparently random series of − and + results, and single measurements on his side cannot make him aware that the distant operator has suddenly changed the orientation of his polarizer. Should we then conclude that there is nothing remarkable in this experiment? To convince the reader of the contrary, I suggest we take the point of view of an external observer, who collects the data from the two distant stations at the end of the experiment, and compares the two series of results. This is what the Innsbruck team has done. Looking at the data a posteriori, they found that the correlation immediately changed as soon as one of the polarizers was switched, without any delay allowing for signal propagation: this reflects quantum non-separability.

Whether non-separability of EPR pairs is a real problem or not is a difficult question to settle. As Richard Feynman once said[16]: "It has not yet become obvious to me that there is no real problem ... I have entertained myself always by squeezing the difficulty of quantum mechanics into a smaller and smaller place, so as to get more and more worried about this particular item. It seems almost ridiculous that you can squeeze it to a numerical question that one thing is bigger than another. But there you are — it is bigger...". Yes, it is bigger by 30 standard deviations. ☐

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Proposal for a loophole-free test of the Bell inequalities

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A proposal for an experimental realization of Bohm's spin-$\frac{1}{2}$ particle version of the Einstein-Podolsky-Rosen experiment is described. Two ^{199}Hg atoms, each with nuclear spin $\frac{1}{2}$, are produced in an entangled state with total nuclear spin zero. Such a state is obtained by dissociation of dimers of the ^{199}Hg$_2$ isotopomer using a spectroscopically selective stimulated Raman process. The measurement of nuclear spin correlations between the two atoms in this entangled state is achieved by detection of the atoms using a spin state selective two-photon excitation-ionization scheme. The experiment will not only close the detector efficiency loophole, but in addition will permit enforcement of the locality condition.

PACS number(s): 03.65.Bz; 32.80.Fb

I. INTRODUCTION

In the early years of the development of quantum mechanics (QM), there was great exhilaration because of its capability of providing accurate statistical predictions. However, in those early days (and even today) there were many who expressed a great deal of concern about the interpretation of quantum mechanics for single microscopic systems. Foremost among them was Albert Einstein, who together with Boris Podolsky and Nathan Rosen (generally referred to as EPR) wrote a paper in 1935 in which they expressed their concern that quantum mechanics was an incomplete theory [1]. Presumably, additional parameters, for which the term "hidden variables" (HV) was coined, would be required in order to restore completeness to the theory. However, since the results of experiments are statistical data, no experiments were immediately obvious and the debate centered on philosophical considerations until 1964.

In that year John Bell published the proof that *any* hidden-variable theory satisfying a physically reasonable condition of locality will yield statistical predictions that must satisfy restrictions for certain correlated phenomena [2]. These restrictions have been derived for various situations and in various forms over the years; all are generally referred to as Bell inequalities. Furthermore Bell demonstrated that quantum mechanics yields statistical predictions that can violate these restrictions. Thus for the first time experimental tests were conceivable.

In 1969 Clauser *et al.* introduced *auxiliary assumptions* to make physically realizable experiments possible with existing technology [3]. Several, involving polarization correlations between two photons in an atomic cascade, were then initiated [4]. The first [5], third [6], and fourth [7,8] gave results in agreement with QM and clearly violated Bell inequalities. Signals observed in the fourth were larger (≈ 1.0 coincidence/sec) than in previous experiments, and systematics could be more thoroughly examined.

In the ensuing years other experiments were performed. In 1982 Aspect, Grangier, and Roger used two channel polarizers and achieved extremely high statistical accuracy [9]. Shortly thereafter, they used time-varying analyzers to change the polarizer settings in a quasiperiodic way [10]. This did not rigorously enforce the locality condition, but was an important advance and has been the only successful progress in this direction. Experiments in recent years have involved correlations between the pair of photons produced by down-conversion in a nonlinear crystal [11–13].

All of these experiments required an auxiliary assumption since they involved photons in or near the visible spectrum and employed detectors whose efficiencies were typically $<20\%$. An inequality that requires no auxiliary assumptions and is therefore called a strong Bell inequality was obtained by Clauser and Horne in 1974 [4,14]; it is especially important because it is formulated in terms consistent with a physically realizable experiment. However, the quantum-mechanical predictions will only violate this inequality for very high detector efficiencies. Strong Bell inequalities have *not* yet been tested experimentally.

The advent of solid-state avalanche photodiodes provides a high detection efficiency that could be sufficient for tests of strong Bell inequalities with photons in or near the visible. Such an experiment has been recently described by Kwiat *et al.* [15].

In this paper we will describe a different type of experimental test of a strong Bell inequality. This experiment also permits enforcement of the locality condition. We will first give a short overview of the experimental concept, then we will briefly discuss the theoretical background, and finally we will present some of the requisite experimental details.

II. EXPERIMENTAL OVERVIEW

An overview of the experiment is shown in Fig. 1. Instead of photon pairs, this experiment involves measurements of the correlations between angular-momentum components of two atoms (nuclei) of the isotope ^{199}Hg [16,17]. The correlated ^{199}Hg atoms are produced by dissociation of ^{199}Hg$_2$ dimers via stimulated Raman excitation to a dissociating state of their $X^1\Sigma_g^+$ ground state. The total electron and the total nuclear spin angular momenta are both zero in the initial rotational state of the mercury dimers, and are not

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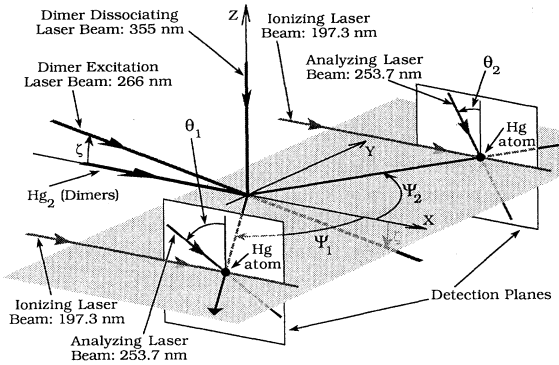

FIG. 1. Schematic of the experiment showing the direction of the mercury dimer beam together with a pair of the dissociated atoms and their respective detection planes. The relative directions of the various laser beams are also shown. Details are discussed throughout the text.

changed in the dissociation process. The two resulting mercury atoms are in $^1S_0$ ground states. Because of the nuclear spin $I=\frac{1}{2}$ of ^{199}Hg, each ground-state atom has total angular momentum $F=\frac{1}{2}$. Consequently we will be observing correlations between components of the spin of two spatially separated spin-$\frac{1}{2}$ particles; it is a direct experimental realization of Bohm's *Gedankenexperiment*. The component of angular momentum (nuclear spin) in any given direction is measured by orienting excitation laser beams in that direction and using polarization-selective excitation of one of the Zeeman sublevels. Atoms with only one component of angular momentum, $M_F=\pm\frac{1}{2}$, are excited. They are then detected ($\approx 100\%$ efficiency) by photoionization via an autoionizing state.

Rates for simultaneous detection (coincidence rates) of the two atoms at their respective detectors are measured for components of the angular momentum in the directions θ_1 and θ_2 (i.e., the directions of the excitation laser beams). A set of four angles can be chosen that give a maximum violation of the strong Bell inequality. This experiment also lays the foundation for an experiment that enforces locality since one can stochastically choose the directions of the excitation laser beams on the nanosecond time scale.

III. THEORETICAL BACKGROUND

Consider a ^{199}Hg$_2$ dimer in an even-J rotational state of the $X^1\Sigma_g^+$ ground state (total electron and total nuclear spin angular momenta are zero; see Sec. IV A 4),

$$|\Psi_T\rangle=|X^1\Sigma_g^+(v,J),S_M=S_1+S_2=O,I_M=I_1+I_2=0\rangle, \tag{1}$$

where S is the electron, and I is the nuclear, spin angular momentum. Subscripts M, 1, and 2 refer to the molecule and the two atoms, respectively. Dissociated atoms are in the 6^1S_0 ground states, for which $S_1=S_2=0$, and the total angular momentum F is the nuclear spin angular momentum I.

The angular momenta can be recoupled so that the spin part of the molecular quantum state is expressed in terms of separated atom basis states $|F,M_F\rangle$,

$$|\Psi\rangle=\sum_{M_{F_1},M_{F_2}}(-1)^{F_1-F_2}\times\begin{pmatrix}F_1 & F_2 & 0\\ M_{F_1} & M_{F_2} & 0\end{pmatrix}|F_1,M_{F_1}\rangle_1|F_2,M_{F_2}\rangle_2. \tag{2}$$

The 3-j symbol couples F_1 and F_2 and their z components to give zero for each sum. For ^{199}Hg we have $F_1=F_2=\frac{1}{2}$, and Eq. (2) becomes

$$|\Psi\rangle=\frac{1}{\sqrt{2}}\{|\tfrac{1}{2},+\tfrac{1}{2}\rangle_1|\tfrac{1}{2},-\tfrac{1}{2}\rangle_2-|\tfrac{1}{2},-\tfrac{1}{2}\rangle_1|\tfrac{1}{2},+\tfrac{1}{2}\rangle_2\}. \tag{3}$$

In terms of two-component eigenspinors, this can be written as

$$|\Psi\rangle=\frac{1}{\sqrt{2}}\left\{\begin{pmatrix}1\\0\end{pmatrix}_1\begin{pmatrix}0\\1\end{pmatrix}_2-\begin{pmatrix}0\\1\end{pmatrix}_1\begin{pmatrix}1\\0\end{pmatrix}_2\right\}. \tag{4}$$

As expected, this is identical to the state of the two spin-$\frac{1}{2}$ particles in Bohm's classic version of the ERP *Gedankenexperiment* [4]. Thus, the spatially separated two-atom system is in an "entangled" state suitable for testing Bell inequalities.

To evaluate the component of angular momentum of one of the atoms in a direction at an angle θ to the Z axis, the rotation matrix for angular momentum $\frac{1}{2}$ that rotates the state through angle θ to the Z axis is required,

$$d^{1/2}(\theta)=\begin{Bmatrix}\cos\frac{\theta}{2} & \sin\frac{\theta}{2}\\ -\sin\frac{\theta}{2} & \cos\frac{\theta}{2}\end{Bmatrix}, \tag{5}$$

together with the projection operator for that component of angular momentum onto the Z axis. The projection operator for $M_F=+\frac{1}{2}$ is

$$P_+=\begin{Bmatrix}1 & 0\\ 0 & 0\end{Bmatrix}. \tag{6}$$

Thus, the probability that $M_F=+\frac{1}{2}$ will be observed in the direction θ is given by the expectation value of the matrix operator,

$$\mathscr{M}_+(\theta)=d^{1/2}(-\theta)P_+d^{1/2}(\theta). \tag{7}$$

We define $R_{i+}(\theta_i)$ to be the rate of detection of atoms (singles rate) with $M_F=+\frac{1}{2}$ in the direction θ_i at detector i, where $i=1$ or 2; similarly, $R_{i-}(\theta_i)$ is the singles rate for $M_F=-\frac{1}{2}$. We define $R_{++}(\theta_1,\theta_2)$ to be the coincidence rate for simultaneous detection of an atom at detector 1 with $M_F=+\frac{1}{2}$ in the direction θ_1 and of an atom at detector 2 with $M_F=+\frac{1}{2}$ in the direction θ_2. Definitions are analogous for $R_{-+}(\theta_1,\theta_2)$, $R_{+-}(\theta_1,\theta_2)$, and $R_{--}(\theta_1,\theta_2)$. Assuming that the total number of dimers dissociating per unit time is N, then the rates for detection of Hg atoms with $M_F=+\frac{1}{2}$ at the two detectors are

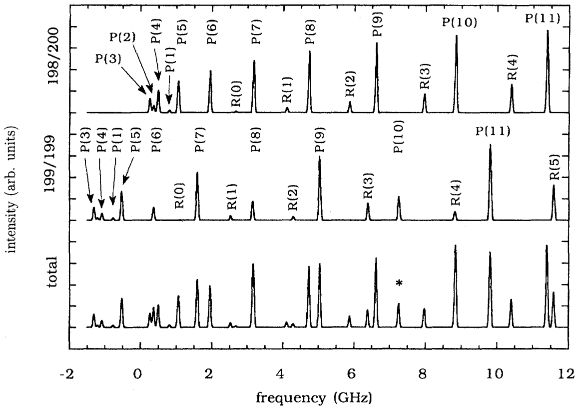

FIG. 3. Simulated rovibrational spectrum for the 58-0 band of the $D\ {}^3\Sigma_u^+ \leftarrow X\ {}^1\Sigma_g^+$ transition for the ^{198}Hg^{200}Hg (top) and the ^{199}Hg$_2$ (middle) isotopomers using natural abundance. Because of the large frequency shift of the ^{196}Hg^{202}Hg transitions, no transitions of this isotopomer are visible in the range of the simulation. As shown in the total spectrum (bottom) the $P(10)$ transition (*) is well resolved. The numbers correspond to the J quantum numbers of the ground state. The simulations show the 3:1 transition intensity alternation due to the spin statistics of the corresponding rotational levels for the ^{199}Hg$_2$ dimer (cf. Sec. IV A 4). A rotational temperature of 3.5 K and a laser linewidth of 60 MHz were assumed.

tions to $P(10)$ of ^{199}Hg$_2$ is $P(9)$ of ^{198}Hg^{200}Hg, which is separated by ≈ 600 MHz. This is much greater than both the laser linewidth and the natural linewidth. The latter depends on the vibrational state and is given by the spontaneous transition probabilities. Even for the largest of these, $A = 7.6 \times 10^6\ \mathrm{sec}^{-1}$ [30,31], the natural linewidth is only 1.2 MHz; i.e., it is much less than the frequency separation of adjacent transitions. In any event, Hg atoms from these other isotopomers would be ignored by the detection system due to both frequency mismatch (Sec. IV B 1) and time of flight (Sec. IV A 6).

Figure 3 shows a simulated rovibrational spectrum for the 58-0 band of the $D^3\Sigma_u^+ \leftarrow X^1\Sigma_g^+$ transition for the two mass 398 isotopomers, ^{199}Hg$_2$ and ^{198}Hg^{200}Hg: natural abundance is assumed. Because of the large frequency shift for the ^{196}Hg^{202}Hg isotopomer, none of its transitions is within the range of the plot; in addition, on the scale of this plot, their intensity would be vanishingly small because of the low abundance of this isotopomer. Only R and P branches are shown since the splitting constants for the three J values corresponding to each N cannot be determined from existing data [18,19]. For the simulations a rotational temperature of 3.5 K and a laser linewidth of 60 MHz were assumed. The simulations show the 3:1 transition intensity alternation due to the spin statistics of the corresponding rotational levels for the ^{199}Hg$_2$ dimer (cf. Sec. IV A 4).

In summary, for a test of the Bell inequality, Eq. (14), the dissociating dimers must have total nuclear spin $I = 0$, a nuclear spin singlet state. Based on the discussion in Sec. IV A 4, only transitions starting with even J can be used. In particular, because of the angular-momentum selection rules for the excitation (266 nm) and stimulated emission (355 nm) transitions, the final dissociating level of the $X^1\Sigma_g^+$ ground state must also have even J and hence zero total nuclear spin. Specifically, the final state J differs from the initial state J by 0, ± 2. The fact that these transitions are electronic singlet-triplet intercombination lines does not alter this conclusion since N must still change by ± 1 in each transition [29]. The $P(10)$ transition is particularly favorable, both because the rotational state population has a peak at $J = 10$ (as discussed in Sec. IV A 3) and because it is well resolved.

6. Conditional detection probability g

The conditional detection probability g of Eq. (8) must be as large as possible [see Eq. (16)]. It is determined by the size of the dissociation volume, and the angular distribution of the dissociating dimer fragments, the size and position of the detectors, and the spread in the velocities of the dissociating fragments.

For the dissociation process the 355-nm laser beam has a diameter of 1.5 mm and is incident along the Z axis (Fig. 1); the 266-nm laser beam has a diameter of 1.0 mm and lies in the X-Z plane at an angle $\zeta \approx 10°$ to the dimer beam. The source volume for the atom pairs is the common intersection of these two laser beams and the supersonic dimer beam. Thus it is a cylinder coaxial with the X axis, with a diameter of 600 μm and a length of 1.5 mm. Both lasers have linear polarizations in the Y direction so as to produce a maximum number of dissociated atom pairs in the directions to the two detectors. Specifically, in the c.m. the atom distribution peaks in the direction of the linear polarization of the lasers. For $J = 10$ the fraction of dissociated atoms in a small solid angle in this direction is a factor $\kappa = 3.8$ greater than if they were isotropically distributed [35].

Momentum conservation requires that, in the c.m. each pair of Hg atoms must have equal and opposite velocities. Furthermore, in the c.m. all Hg atoms produced by the dissociation process have essentially the same speed. The spread in their c.m. speeds is determined by the very narrow frequency spread of the dissociating lasers. Their directions are spread over 4π Sr in the c.m. with peaks in the distribution in the direction of the linear polarization of the dissociating lasers [35].

However, for a given direction Θ in the c.m. the directions Ψ_1, Ψ_2 and the velocities V_1, V_2 in the laboratory frame are determined by vector addition of the dimer velocity V_0 with the c.m. velocity of the corresponding Hg atoms (cf. Fig. 4). The detectors and apertures are positioned at these laboratory angles.

With the detector geometry fixed at these angles, it is clear that the smaller the spread in the velocities of the Hg$_2$ dimers the higher the conditional probability. Based on the mean speed $V_0 = 412$ m/sec and the estimated speed ratio $S = 36$ in the supersonic expansion, the spread in dimer velocities is $\Delta V \approx 19$ m/sec, FWHM. For this velocity spread, optimum detector positions, and a usable source size, the conditional probability in Eq. (16) is $g < 0.9$.

To obtain larger values of g, the Doppler effect in the transition step at 266 nm will be used to spectroscopically dissociate only dimers within a velocity spread $\Delta V = 3$

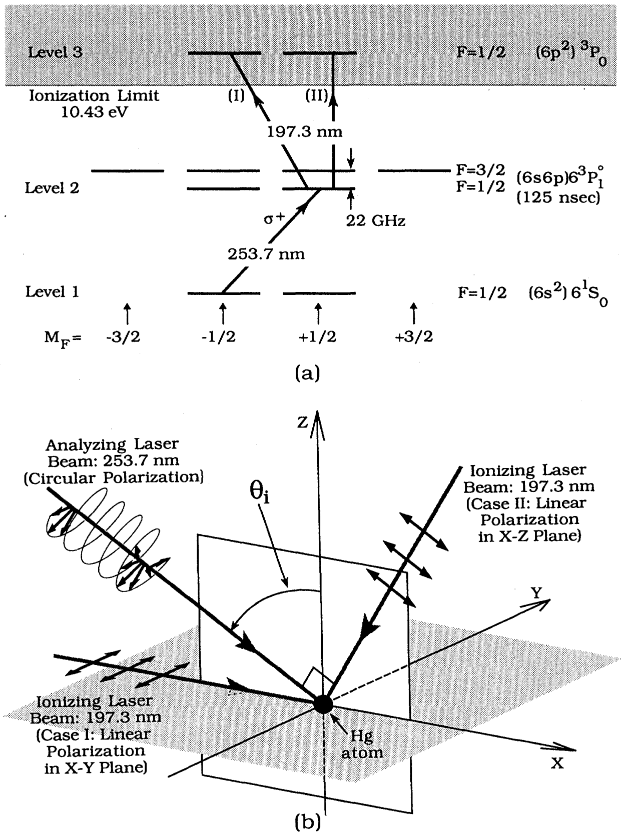

FIG. 6. (a) Relevant energy levels of the Hg atom and the corresponding transitions for detection. This example shows an angular-momentum analysis using right circularly polarized light. Since M_F must increase by 1, only ground-state atoms with $M_F = -\frac{1}{2}$ can be excited to the $6^3P_1^o$ $(F=\frac{1}{2})$ state. Cases I and II correspond to different laser alignment and polarization schemes for the transition to the autoionizing state, as shown schematically in (b).

dissociation angles and points of origin of the fragments within the source volume.

B. Mercury atom detection

As discussed in Sec. III, the highest possible efficiency for detecting both of the Hg atoms from each dimer is required to test the strong Bell inequalities [cf. Eq. (16)]. High detection efficiency η is achieved by using a two-step excitation-ionization process. Immediately following the entrance aperture to each detector (see Fig. 1), the Hg atoms pass through two laser beams with wavelengths of 253.7 and 197.3 nm. As shown in Fig. 6(a), the first laser is circularly polarized and drives the transition from the $(6s^2)6^1S_0$ $(F=\frac{1}{2})$ ground state (level 1) to the $(6s6p)6^3P_1^o$ $(F=\frac{1}{2})$ state (level 2). The second laser drives the transition from level 2 to the $(6p^2)^3P_0$ autoionizing state (level 3); two cases for its polarization and orientation will be considered [cf. Fig. 6(b)]. At the point of ionization the Hg^+ energy is 0.76 eV (for symmetric dissociation velocities in the laboratory frame) and the photoelectron energy is 0.74 eV. Atom detection is via both the resulting ion and the photoelectron.

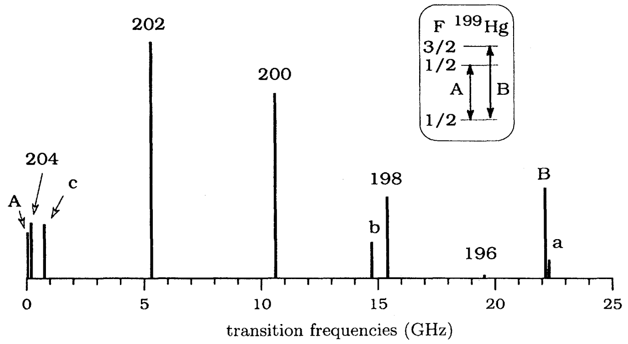

FIG. 7. $6^1S_0 \rightarrow 6^3P_1^o$ transition frequencies for Hg isotopes relative to the desired ^{199}Hg transition (labeled A). Lowercase letters indicate three of the transitions in ^{201}Hg. The isotope ^{204}Hg is suppressed in the dissociation step (cf. Sec. IV A 5).

1. Atomic excitation rates

From the measured value of the spontaneous emission probability A_2 for the $(6s6p)6^3P_1^o$ state (equivalently its lifetime), the induced absorption probability per unit time R_{12} for the 253.7 nm excitation transition can be evaluated [36],

$$R_{12} = \frac{g_2}{g_1}\frac{\lambda^3}{8\pi hc}\mathscr{I}_1(\nu)A_2 . \tag{21}$$

Here $\mathscr{I}_1(\nu)$ is the spectral intensity (power per unit area per unit frequency interval $d\nu$), and g_i is the statistical weight of state i. Using $A_2 = 8\times 10^6\ \mathrm{sec}^{-1}$, [37] we get

$$R_{12} = 7.85\times 10^{14}\mathscr{I}_1\ \mathrm{sec}^{-1} , \tag{22}$$

where $\mathscr{I}_1$ is the 253.7 nm intensity in units of $\mathrm{W\,cm^{-2}\,Hz^{-1}}$. The $6^1S_0 \rightarrow 6^3P_1^o$ transition frequencies for Hg isotopes, with respect to the relevant transition of ^{199}Hg (labeled A), are shown in Fig. 7 [38].

2. Atomic ionization rates

Two cases for polarization and orientation of the ionizing laser are considered. Cases I and II in Figs. 6(a) and (b). For case I the ionizing laser beam propagates on the X axis and is linearly polarized along the Y axis. For these conditions, the ionization transition probability is independent of the angle θ of the excitation laser beam, and all of the Zeeman sublevels of the $F=\frac{1}{2}$ and $F=\frac{3}{2}$ states of $6^3P_1^o$ can be driven to the autoionizing $(6p^2)^3P_0$ state. For case II the ionizing laser beam propagates in the X-Z plane at right angles to the excitation laser beam; it is linearly polarized in the X-Z plane (i.e., parallel to the propagation direction of the excitation laser). In this case the selection rule is $\Delta m_F = 0$; consequently, atoms in the Zeeman sublevels $m_F = \pm\frac{3}{2}$ of $6^3P_1^o$ cannot be excited to the autoionizing level. In practice, case II is more difficult to implement since the ionizing and excitation laser beams must be kept perpendicular as θ is varied.

Determination of the average transition probability to the autoionizing state $(6p^2)^3P_0$ is made by using the measured width, $\Gamma_3 = 9\ \mathrm{cm}^{-1}$, [39] of this state together with a calculated value, $f_{23} = 0.362$, [40] for the oscillator strength of the

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High-temperature high-pressure all-metal pulsed source of van der Waals dimers: Towards the Einstein-Podolsky-Rosen experiment

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An all-metal pulsed source of van der Waals (vdW) dimers was constructed; it operates at temperatures up to 1000 K and carrier gas stagnation pressures up to 10 bars. Performance of the source was demonstrated in the production and spectroscopy of both CdAr and Cd_2 molecules in a supersonic beam expansion. Simulation of the recorded laser induced fluorescence (LIF) excitation spectra using the $B^3 1(5^3P_1) \leftarrow X^1 0^+(5^1S_0)$ and $b^3 0_u^+(5^3P_1) \leftarrow X^1 0_g^+(5^1S_0)$ transitions in CdAr and Cd_2, respectively, showed that these molecules were produced with a rotational temperature in the range from 3 K to 19 K. The source was incorporated into an experimental set-up dedicated to the realization of Bohm's spin-1/2 particle version of the Einstein-Podolsky-Rosen experiment for $(^{111}Cd)_2$ molecules. *© 2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4747274]

I. INTRODUCTION

In molecular beam experiments pulsed supersonic sources are generally preferred over those operated in a continuous mode because (i) they minimize consumption of the carrier gas, usually a noble gas (Ng), and (ii) because they require considerably less pumping capacity. Furthermore, with a pulsed beam expansion, sufficient cooling conditions[1] can be achieved using a larger nozzle diameter and a lower Ng stagnation pressure. Consequently, a pulsed valve offers higher transient beam densities while reducing the average Ng load on the pumping system. In addition, a pulsed molecular beam matches well with a pulsed laser, and with a pulsed detection and data acquisition system. The main disadvantage of a pulsed valve is the limited ability to operate it at a high temperature (commercially used solenoid valves cannot be operated above 590 K),[2,3] an option which has been common for continuous beam sources. In the literature one can only find a few examples of pulsed molecular beam sources operating at relatively high temperatures. Bahat *et al.*[4] described a pulsed conical stainless-steel nozzle source for aniline or zinc tetrabenzoporphyrin large clusters with low rotational temperatures (3–15 K). It operated at temperatures up to 740 K and carrier gas stagnation pressures up to 3 bars; it had 0.3–0.6 mm changeable orifices. It was possible to heat the solenoid valve because it used a glass fiber-insulated solenoid coil and Kapton® seals. Excessive heating of the solenoid valve could be avoided to some extent by using a longer plunger and by cooling the solenoid itself while heating the orifice to the desired temperatures. Using this approach Li and Lubman[5] introduced a high-temperature pulsed solenoid valve with a 0.8 mm orifice that operated at temperatures up to 820 K and at a 1.3 bars stagnation pressure; it was tested with several organic molecules. A similar pulsed valve was used by Wang and Li[6] to combine flow injection analysis with multiphoton ionization time-of-flight mass spectrometry. A continuous-purge pulsed valve for high-temperature applications was reported by Senkan and Deskin.[7] Their valve had a 0.5-mm orifice and was tested for benzene; it provided stable operation up to 770 K at 0.3 bar stagnation pressure. Fink *et al.*[8] constructed a high-temperature pulsed supersonic nozzle made of quartz with a 0.1 mm orifice. It operated in the temperature range 900–1300 K and at carrier gas stagnation pressures up to 2 bars. It was used for production of NO molecules with high rotational temperatures (50–700 K).[9] Very recently, Shen and Sulkes[10] reported a high-temperature pulsed solenoid valve with long-term operation at 670 K in a time-of-flight molecular beam system.

In supersonic beam sources employed for production of CdNg and Cd_2 molecules, it is essential to heat the Cd sample and the nozzle to temperatures well above the Cd melting point (594 K). Most previous experiments with CdNg and Cd_2 expansion beams either employed continuous sources operated at temperatures 900–950 K (Refs. 11–15) or used pulsed Nd:YAG-laser-vaporized Cd samples.[16] The later required an additional Nd:YAG vaporization laser and a special intra-source mechanism dedicated to the rotation and translation of the Cd rod; this vaporization process also produced additional uncontrollable excitation of the CdNg molecules from their ground to low-lying electronic energy states. Finally, Okunishi *et al.*[17] described the single example of a high-temperature pulsed valve used for the spectroscopy of CdAr molecules. The valve operated at temperatures up to 873 K and at carrier gas stagnation pressures up to 4 bars; it had a 0.2 mm orifice. The 100-mm long plunger was attached to a water-cooled commercial fuel injector; it operated at a 10-Hz repetition rate with 2–3-ms wide pulses.

In this article we describe an all-metal, high-temperature, high-pressure pulsed source of CdNg and Cd_2 van der Waals (vdW) complexes. It has interchangeable cartridges so that different orifice diameters (D) in the 0.06–0.5 mm range can be used. This pulsed source operates at temperatures up to 1000 K and stagnation carrier gas pressures (p) up to 10 bars. It has a large (15.2×10^3 mm^3) reservoir for Cd. Performance of the source was demonstrated via the

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