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Evident atoms: visuality in Jean Perrin's Brownian motion research

Charlotte Bigg

Max Planck Institut für Wissenschaftsgeschichte, Boltzmannstrasse 22, 14195 Berlin, Germany

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ABSTRACT

The issue of shifting scales between the microscopic and the macroscopic dimensions is a recurrent one in the history of science, and in particular the history of microscopy. But it took on new dimensions in the context of early twentieth-century microphysics, with the progressive realisation that the physical laws governing the macroscopic world were not always adequate for describing the sub-microscopic one. The paper focuses on the researches of Jean Perrin in the 1900s, in particular his use of Brownian motion to produce evidence of the existence of atoms and in favour of the kinetic theory. His results were described by many contemporaries, and subsequently by historians, as the first direct proof of atomic and molecular reality. The paper examines the different strategies developed by Perrin for bridging the macro and sub-microphysical realms and making the latter accessible to the senses; even though neither atoms nor molecules were ever actually seen, and in fact very few visual representations were shown and published in connection with these experiments. This case provides a good example of how visualizing, representing and convincing could be interwoven in the production of evidence about the sub-microphysical realm circa 1900.

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1. Introduction

At the turn of the twentieth century the physical sciences became increasingly concerned with sub-microscopic entities, including atoms, molecules, electrons and ions. Scientists in different fields groped in the sub-microscopic dimension searching for ways of perceiving, studying and measuring these problematic entities. They devised a number of indirect means of showing the existence and of studying the properties of these new and imperceptible objects. Some of the tools of exploring the microscosmos, for example to track the electron, include cloud chambers, oil drops and vacuum tubes. The recurrent issue of shifting scales between the microscopic and the macroscopic dimensions thereby acquired new meanings at the turn of the twentieth century.[1] In connection notably with the investigation of radioactivity, X-rays, and of a spate of new particles, the realisation progressively emerged that the physical laws governing the macroscopic world were not always adequate for describing the sub-microscopic one.

This paper focusses on the researches of Jean Perrin in the 1900s, in particular his use of Brownian motion to produce evidence of the existence of atoms and in favour of the kinetic theory. His results were described by many contemporaries, and subsequently by historians, as the first direct proof of atomic and molecular reality. Thus Perrin was awarded the physics Nobel prize in 1926 for having 'put a definite end to the long struggle regarding the real existence of molecules'.[2] This arguably set the tone for a series of visualizations of the molecule and the atom produced throughout the twentieth century using innovative technologies, down to the scanning tunnelling microscope in the 1980s.[3] This paper examines the different strategies developed by Perrin for bridging the macro and sub-microphysical

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E-mail address: bigg@mpiwg-berlin.mpg.de

[1] Discussions of the issue of shifting scales in earlier times include Sibum (2001); Shickore (2003). It is also of course a matter of much interest to philosophers of science. See, for example, Hacking (1983).

[2] Physics Nobel Prize Presentation Speech by Prof. C. W. Oseen, 10 December 1926 (*Nobel Lectures, Physics 1922–1941*, 1965). Note that though Perrin distinguished carefully between molecules and atoms in his scientific argumentation, he put forward his work as proving the reality of both interchangeably, as a consequence of the particulate nature of matter.

[3] See, for example, Hennig (2004).

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realms and making the latter accessible to the senses; even though neither atoms nor molecules were ever actually seen, and in fact very visual representations were shown and published in connection with these experiments. This instance provides a good example of how visualizing, representing and convincing could be interwoven in the production of evidence about the sub-microphysical realm circa 1900—*evidence* being a particularly appropriate locution here, whose etymology (*e-videre*) already encapsulates the link between visuality and proof.

Perrin's Brownian motion work has been thoroughly investigated by historians, most notably by Mary-Jo Nye and Stephen G. Brush.[4] It has also been a favourite case for philosophers' discussions of realism, the nature of scientific evidence and of scientific explanation.[5] This paper complements these sophisticated analyses of Perrin's argumentation and of its significance for the history of the physical sciences by focussing specifically on the importance of *visuality* in Perrin's Brownian motion work, and more widely in his programme of scientific, disciplinary and institutional renewal. I am interested here not only in the role played by visuality in Perrin's evidence of the existence of atoms but also in grasping the epistemological, rhetorical and instrumental components of Perrin's particular sense of 'molecular reality' in the early years of the twentieth century. By making a close reading of his scientific and popular writings and bringing in new sources that throw light on his training and local interactions, the paper also emphasizes the coherence and autonomous nature of Perrin's research above and beyond the experimental confirmation of Albert Einstein's theory of Brownian motion in 1905.

2. Recasting the atomic debate, elaborating French physical chemistry

The elementary constitution of matter, whether it was continuous or discontinuous, was subject to notorious controversies throughout the nineteenth century. Since Dalton's claim in 1808 that a particular type of particle corresponds to each chemical element, chemists debated whether atoms were perceptible entities or rather fictitious constructions, working tools, mere hypotheses that did not correspond to any tangible reality. The fixed proportions of substances involved in chemical reactions were referred to with some vagueness as 'atoms', 'equivalents' or 'volumes'. More empirically inclined chemists refused to presume the existence of entities that had not and, they argued, could not be seen. Others, pointing to the fruitfulness of many laws and theories involving atoms, in particular the kinetic theory of gases, argued that their existence should not be excluded *a priori*.[6]

At the root of these confrontations lay a debate about scientific method, in particular about the use of mechanical models. Chemists understood these to be entities underlying mechanical laws, for example vortices or atoms, that helped account for macroscopic phenomena, such as the diffusion of liquids across membranes. Chemists associated the expression 'mechanical model' with atomism and with physical reductionism, the belief that chemistry was a more superficial and less effective approach than physics. This view was espoused by some physicists but it was mostly discussed by chemists fearing such a development. In the decades around 1900, chemists actively discussed the opportunity of adopting physical instruments, methods and concepts into their field. While chemists favourable to these methods such as Perrin developed the field of physical chemistry others remained sceptical. The issue of atomic reality was inextricably embedded in these debates and in the rise of modern physical chemistry.[7]

From the mid-nineteenth century and the development of thermodynamics, atomism, or rather as they usually put it, the discontinuity of matter, was also a matter of debate among physicists, because the strongest argument in favour of atomism, the kinetic theory, appeared to be in part incompatible with the second law of thermodynamics. James Clerk Maxwell and Ludwig Boltzmann's proposals to bring the law of entropy in agreement with the kinetic theory by considering this law no longer as absolutely but as statistically valid, was problematic for many physicists. On the other hand, the fruitfulness of the kinetic theory in accounting for many phenomena, together with the discovery of a number of subatomic particles around 1900 gave physicists good reasons to embrace atoms. Perrin sought to address these issues in all their different, epistemological, theoretical, experimental, physical and chemical dimensions. His strategy, I will suggest, was to shift the terms of the debate about atomism and transform it into a pragmatic issue that might be clarified by experiments. Simultaneously he attempted to transcend the conflicts by proposing a new basis for physical chemistry.

In 1898, shortly after completing his studies in physics and chemistry at the prestigious Ecole Normale Supérieure with a Ph.D. showing that cathode rays are negatively charged, Perrin was put in charge of developing a new course in physical chemistry at the Sorbonne. From 1900 he also gave lectures on thermodynamics, optics and acoustics at the Ecole Normale Supérieure.[8] Simultaneously, he began investigations towards the establishment of a laboratory for physical chemistry at the Sorbonne, travelling abroad[9] to examine laboratories, probably visiting J. J. Thomson in Cambridge (where his friend the physicist Paul Langevin was staying), Wilhelm Ostwald in Leipzig, and Jacobus H. Van t'Hoff in Berlin. While the first's work had been influential on Perrin's first investigations (cathode rays, X-rays, ions), the latter two were foremost representatives of physical chemistry in Europe.[10] As his writings and public lectures in the early 1900s testify, Perrin was also becoming increasingly involved in the chemists' debate about the atomic hypothesis;[11] while he was led through his teaching to reflect on thermodynamics and its relation to mechanics.[12]

Many of the strategies and ambitions expressed in Perrin's Brownian motion publications and later were developed in these years. Perrin's first book *Traité de chimie physique. Les principes*, published in 1903, is an attempt to lay out systematically the

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[4] For a comprehensive analysis of Perrin's Brownian motion research and its immediate reception, see Nye (1972). Brush (1968) gives a detailed survey of the history of Brownian motion research from the early nineteenth to the early twentieth century. See also Maiocchi (1990).

[5] For example Salmon (1998), pp. 86–91 (argument presented orally in 1980); Cartwright (1983), pp. 82–86; Mayo (1988); Achinstein, (2001), pp. 243–265; Achinstein (2002). I am grateful to Philip Kitcher for pointing me to this literature.

[6] Nye (1972), pp. 1–50; Klein (2004).

[7] On chemists' reluctance to use what they perceived to be physics-based approaches and physical instrumentation such as spectroscopes, see Bigg (2002), Part II.

[8] Aimé Cotton, untitled typescript in memory of P. Langevin and J. Perrin, 1936, Fonds Jean Perrin 54J, Archives de l'Académie des Sciences, Paris (hereafter Arch. Acad. Sci., Paris).

[9] Perrin (1918), p. 25.

[10] In a letter to Paul Langevin then in Cambridge (undated, early 1898), Jean Perrin wrote: 'Pour mon voyage ... j'aurais bien envie d'aller d'abord à Cambridge, puis de gagner Leipzig (Ostwald) et Berlin (Van t'Hoff) par Anvers et Cologne. Si tu crois vraiment que cela soit bon, dis-le moi j'écrirai au Prof. J. J. Thomson. Outre le plaisir que j'aurais à causer avec lui, la joie que j'aurais à te voir il me semble qu'il ne serait pas inutile de voir un laboratoire anglais bien monté avec un guide ... comme toi' (Box 76, Fonds Paul Langevin, Centre de Ressources Historiques, Ecole de Physique Chimie Industrielles de la Ville de Paris).

[11] Perrin (1901, 1906a).

[12] Perrin (1923), p. 20.

fundamentals of physical chemistry. It is also a profession of faith in atomism and the value of hypothesis in science. But rather than explicitly taking sides in the ongoing disputes about atomism—at the time the anti-atomist stance was pervasive among French physical scientists outside the Ecole Normale Supérieure—Perrin proposed to overcome it entirely.

In the book's introduction, Perrin first described both positions as following: while thermodynamics had been built up using scientists' faculties of analogy and generalisation (the inductive method); atomistics had been developed using scientists' powers of imagination (the deductive, or intuitive method), 'that consists in imagining *a priori* matter to have a structure that still escapes our imperfect senses, such that its knowledge would by deduction enable predictions to be made about the perceptible properties of the universe'.[13] Perrin acknowledged that the first had rightly been suspicious of the second, who systematically privileged 'explanations of the visible by the invisible, even when they did not lead to the discovery of new facts'.[14] And indeed, Perrin's book, he claimed, was entirely dedicated to the applications of this inductive method. Yet he could not endorse blanket condemnations of 'molecular theories'. A reasoned critique of these and of the deductive method was possible, that preserved their very essence, that is, 'the search for a conception attributing to matter a structure from which its perceptible properties might easily be deduced'.[15] Perrin claimed that thereby 'I do not fall back into metaphysics. I do not forget that sensation is the only reality'. But there was a crucial proviso: 'It is the only reality, provided that to actual sensations all *possible* sensations are added'.

By analogy with microbes, Perrin put forward atoms as belonging not the realm of metaphysics but to that of possible sensation:

> Pasteurian theory might have been entirely developed without the microscope, from the premise that contageous diseases resulted from the multiplication of very small living beings . . . who can, from there, seriously sustain that the domain of possible sensations cannot exceed the actual domain?[16]

Likewise the scientists who attributed a granular structure to apparently homogeneous matter.[17] For Perrin, the fruitfulness of the molecular theory should be acknowledged, and the debate between energeticists and atomists, between inductivists and deductivists, and about the value of hypothesis in science should be brought to a close by the 'conciliation of two methods that are by no means incompatible'.[18]

Among the many attempts to conceptualise and overcome the conflicts about atomism in this period, Perrin's book is noteworthy in several respects. For the purposes at hand, we need insist especially on the way in which Perrin sought to concile the empiricist emphasis on the role of the senses in scientific investigation with the use of hypotheses; and his appeal to microbiology as an epistemological model for connecting the macro and micro worlds. Underlying this original approach was the conviction that physical science was in need of clarification. Perrin's book was put forward as an attempt to reformulate the fundamental principles of physical science such that they always derived from the sensory, experimental perception of the world:

> I have collected in this first book the principles whose study and discussion constitute a natural introduction to the different physical sciences. These principles' similarity derives not only from their extreme generality, and hence their broad philosophical implications, but also because each of them can be suggested by the comparison of known facts, without making it necessary to represent the world other than it appears to us. In all of them the same slow and general march of the particular towards the general can be found, the same distrust of mystery and metaphysics, the same contempt for all that cannot be reduced to *actually realisable sensation*.[19]

Perrin further advocated the 'reform' of tainted language, 'ways of speaking that long-vanished beliefs had imposed, and that too often make language unintelligible'. Some words, such as 'induction' and 'deduction', were like worn coins on which the effigy was no longer recognisable.[20] Confusion over the definition of such terms as 'force' or 'energy', had led the energeticists to move away from 'physical reality' and turn science into a form of religious teaching. Perrin argued for new habits of precision in language and thinking, a recommendation with unmistakable moral connotations,

> for there is not in us an intellectual and a moral being . . . ignoring each other . . . Him who has once clearly recognised through analysis that a sentence can be devoid of meaning and without foundation . . . will perhaps refuse to subject his actions to cruel or absurd conventions, even when these conventions have been established by authority or tradition.[21]

In the typical style of the Third Republic's progressive humanism, Perrin affirmed his faith in the power of reason to overcome confusion and disputes; while advocating a very personal brand of empiricist atomism.[22]

Adressing another long-standing tension in the physical sciences Perrin also discussed the relationship between physics and chemistry and the nature of physical chemistry. Physics, for Perrin, was peculiarly concerned with continuous phenomena (continuous variations, equilibrium, reversibility) while chemistry dealt with discontinous ones (discrete elements, irreversibility). Accordingly, physics privileged mathematical analysis (continuous functions) and precision measurement, while chemistry multiplied the tools for bringing different bodies into contact and into different conditions of pressure or temperature. Physical chemistry for Perrin combined the theoretical and instrumental ressources of both approaches to investigate phenomena neglected by both parties, such as reversible chemical transformations (using the ressources of physics), or irreversible physical processes (using chemical techniques). Here also, Perrin strove for a synthesis, one that would bind more closely together physics, chemistry and mathematics. Physical chemistry, according to him, was made up of three major branches: the science of chemical equilibria, or chemical statics; the science of irreversible transformations, or chemical dynamics; and stoechiometry, the study of the influence of the composition of bodies on their properties.

Perrin's attempt to recast the foundations of physical science and more specifically, of physical chemistry appear in retrospect somewhat idiosyncratic and does not seem to have gained much

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[13] Perrin (1903), p. viii; all translations are my own unless otherwise specified.

[14] Ibid.

[15] Ibid, p. ix.

[16] Ibid., pp. ix–x.

[17] Ibid., p. x.

[18] Ibid.

[19] Ibid., p. viii; original emphasis.

[20] Ibid., p. xii; Perrin (1906b), p. 81.

[21] Ibid., p. xvi.

[22] On Perrin's (left-wing) political outlook on science and society, see Nye (1975).

of a following (for example his proposal to rename the second principle of thermodynamics the ‘principle of evolution’) but it does provide some useful insights into his conceptions of science and his perceived role within it. That Perrin, in his very first book could pretend to reform physical science is certainly not unrelated to the elite status of the Ecole Normale in the French scientific landscape and the self-confidence of its alumni; but also to the expectations placed in him by the authorities in the French physics of the time.

In his report on Jean Perrin’s work and evaluation of his suitability to take over the teaching of physical chemistry at the Sorbonne, Henri Poincaré argued in 1898 that Perrin, despite his young age, lack of experience and of training in physical chemistry, presented all the qualities required to establish this new field in the country:

> Abroad, hardy pioneers have charged ahead, preempting experimentation and worrying little that it had trouble following. They have arrived at a set of conceptions that are extremely plausible but yet to be verified. Excepting a few great names of whom France can justly be proud, these innovators have found here few imitators or disciples. It seems as if the French mind avid of clarity and logic, is reluctant to embark on such reckless adventures. The professor to be appointed will have to introduce us to the work of these scientists and naturalise it here while clarifying it. He will need a mind open enough to study them with sympathy and understand them, but independent enough not to follow them blindly. He will need a little of what they have and some of what they do not, in particular a taste for precision.[23]

Poincaré further pointed out that the new professor would have to be able to discern simple laws beyond the complexity of phenomena, and that Perrin seemed to present the qualities required. Perrin was elected with a small advance over his main competitor, his friend, the well-established chemist Pierre Curie. These expectations help explain how Perrin, who had until then done work very much in the line of Cavendish physics, could now attempt to elaborate a specifically French style of physical chemistry in the early 1900s.

This constituted the programmatic scientific and epistemological framework within which Perrin launched into his Brownian motion experiments. The following sections examine how Perrin applied this reasoning to one specific hypothesis, the atom, and sought to confound the stance that atoms were fundamentally inaccessible to the senses by devising an experiment that brought them within the realm of possible, if not actual sensation.

3. Jean Perrin’s Brownian motion investigations

Histories of the scientific investigation of Brownian motion have varied little since they first appeared in the 1910s. According to these, Brownian motion, the perpetual and irregular motion of particles suspedended in a liquid or a gas, was acknowledged for the first time in print by the botanist Robert Brown in 1827. In the following decades researchers established that this motion was due neither to organic processes or to the liquid’s evaporation, nor to convection.[24] Circa 1990 Brownian motion was put forward as a decisive criterion to test the validity of the kinetic and the thermodynamic theories. In the introduction of his 1905 article on the subject Albert Einstein stated:

> If this motion with all its expected regularities is really to be observed, then classical thermodynamics cannot be seen as absolutely valid at the microscopic level, and then an exact determination of atomic size is possible. Conversely, should the prediction concerning this motion be unfounded, then a grave argument against the molecular–kinetic concept of heat will have been provided.[25]

It was Einstein who most forcefully set up Brownian motion as a crucial phenomenon whose analysis would decide on the validity of *either* atomism *or* classical thermodynamics, and, most importantly, he provided new quantitative methods for apprehending the problem. Developing strands from his doctoral thesis, Einstein proposed a new method for determining molecular dimensions and Avogadro’s number N using diffusion theory and hydrodynamics; and he proposed new methods for measuring the irregular motion of individual particles in solutions.[26]

Einstein was however not the first to express this view, as historians have remarked. The French physicist Léon Gouy had made similar claims in 1895 (albeit without providing quantitative methods). Gouy, rather than Einstein arguably provided the initial impetus for Jean Perrin as he embarked on his study of Brownian motion. In paper of 1904 Perrin thus argued that Brownian motion supplied evidence of the statistical nature of the second principle of thermodynamics.[27]

In 1903, his laboratory freshly, if not lavishly fitted out, Perrin was ready to undertake experimental work again, and he began investigating colloid solutions,[28] his experiments geared to address the issue of atomism, culminating in his major paper of 1909, ‘Mouvement brownien et réalité moléculaire’.[29] Perrin’s arguments have been the subject of thorough analyses which do not need to be repeated here.[30] I discuss here only the features most pertinent for the purposes at hand.

Perrin deployed a range of methods to show that established theories on the behaviour of gases and solutions were applicable to colloid solutions, and, at the same time, that mechanical theories developed for macroscopic phenomena could be extended to suspended colloid particles. For example, Perrin argued that Avogadro’s Law (stating that two gases at the same conditions of temperature and pressure contain the same number of molecules) implied that molecular energy was also a universal constant.[31] Perrin then pointed out that van’t Hoff had extended this invariance of molecular energy to molecules in all diluted solutions. Perrin speculated that it was possibly true of all liquids, and even of minute particles suspended in a solution. If one could observe and measure the molecular energy of such particles, he argued, it would be possible to establish whether solutions of suspended particles obeyed the same laws as gases.

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[23] Henri Poincaré, *Rapport sur la candidature de Mr. Perrin à la chaire de chimie physique*, undated, ca. 1898, Jean Perrin file F/17/24822, Archives Nationales, Paris.

[24] See Brush (1968).

[25] Einstein (1905), p. 549.

[26] For a detailed analysis of Einstein’s argument, see Stachel (1989), pp. 41–55, 170–182, 206–222. On the place of these investigations in Einstein’s pursuits in the 1900s and his ‘interdisciplinary atomism’, see Renn (1997). See also Klein (1967, 1982); Renn (2005).

[27] ‘si l’on pouvait contester l’existence des molécules, on ne peut contester l’existence du mouvement brownien, et ce n’est plus une fiction que d’imaginer un systeme assez petit, assez élémentaire, pour que le principe de Carnot ne puisse lui être appliqué sans précaution’ (Perrin, 1904–1905, reprinted in Perrin, 1951, p. 122; Gouy 1895).

[28] Perrin (1923), p. 20.

[29] Perrin (1909).

[30] See Nye (1972); Brush (1968); Achinstein (2001, 2002).

[31] The pressure of a gas on its container, limiting its expansion, is explained by the collision of molecules such that $pv = \frac{2}{3}nw$ (p being the pressure exercised by n molecules, of mean kinetic energy w in volume v). If n = N, then pv = TR (T absolute temperature, R perfect gases constant), so $\frac{2}{3}Nw = RT$, and since N is the same for all bodies, the mean molecular kinetic energy of translation has the same average value for all gases: $w = \alpha T$. So α, the molecular energy constant, is, like N, a universal constant.

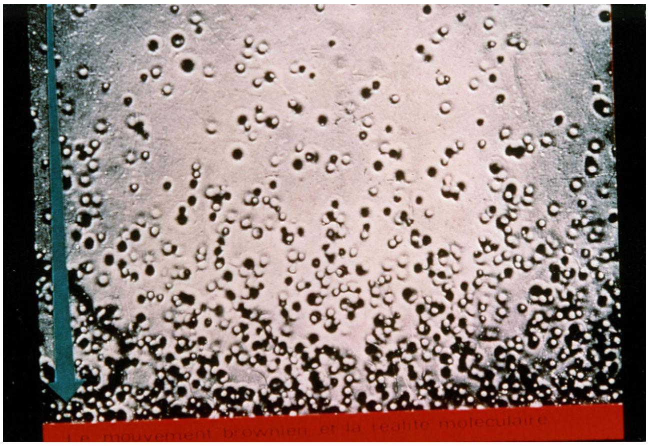
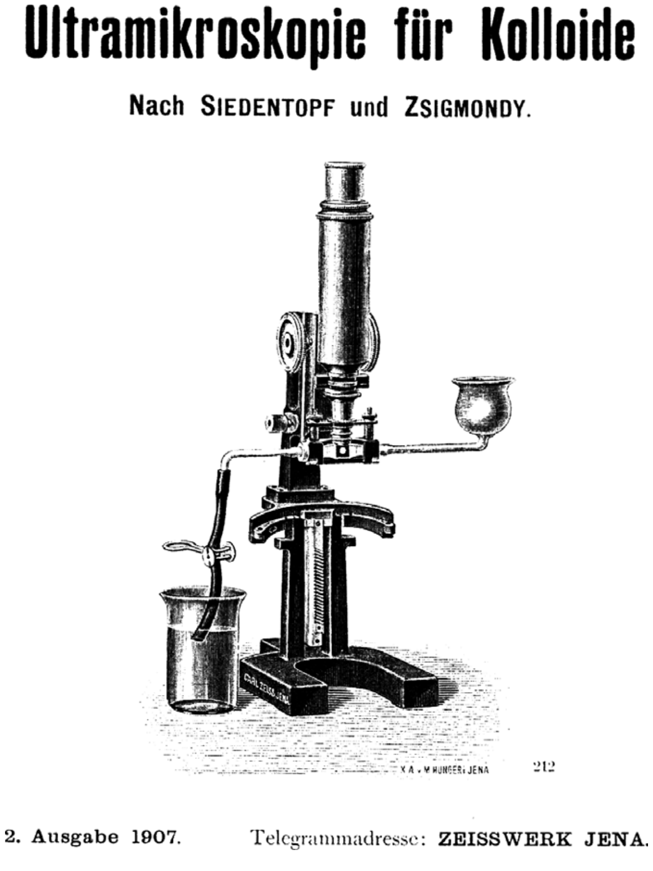


Fig. 1. A microphotography of the height distribution of particles of resin suspended in water. This photograph was taken by Jean Perrin and subsequently exhibited in the science museum he founded in Paris in 1937, the Palais de la Découverte (© Palais de la Découverte, Paris).


Ultramikroskopie für Kolloide

Nach SIEDENTOPF und ZSIGMONDY.

2. Ausgabe 1907. Telegrammadresse: **ZEISSWERK JENA.**

Fig. 2. One of the early ultramicroscope price lists issued by Zeiss. The light-producing apparatus is not shown (© Zeiss Archiv, Jena).

In order to test this hypothesis, Perrin prepared an emulsion of gamboge grains of the same size. When the emulsion was left at rest for a while, the grains settled such that their concentration diminished exponentially with height (see Fig. 1). Perrin set out to show that the distribution equation, the ratio of grain concentrations between two levels of the solution, was identical to that describing the concentration of gases in the atmosphere, as predicted by the kinetic theory. For this it was necessary to determine the density of the grains, as well as their number and radius.[32] Perrin used these measurements to calculate a value of the grain energy of his emulsion, which proved equal to the average energy of a molecule at the same temperature. Using three different methods, Perrin obtained values for the molecular energy of the colloid solution which all agreed with each other, and with the value predicted by the kinetic theory of gases.[33] He came to the conclusion that

> The origin of Brownian Motion can be seen as experimentally confirmed and simultaneously it makes a denial of the objective reality of molecules difficult to sustain. Brownian motion supplies us on a different scale a true picture of the motion of molecules.[34]

In the second part of his article, Perrin made use of Einstein's method of determining the average displacement energy of individual colloid particles.[35] He ended his paper with a list of indepedent methods, developed by himself or others, of determining N, concluding that all these methods led to one and only value:

> The extreme diversity of phenomena which lead to the same numerical value make resistance to the molecular hypothesis difficult to sustain.[36]

Perrin's argument rested essentially on a demonstration that colloid solutions bridged the macroscopic and microscopic dimensions, in that they functioned according to the laws governing both: he showed that the kinetic theory of gases could be applied to suspended particles; but also that Stokes's law (describing the movement of a sphere in the ether), was valid for minute particles. He was able to do so by establishing what he called a 'concordance', or agreement between the values obtained using a number of different methods; but also the agreement of values obtained through experimental and theoretical means. On the basis of this concordance, Perrin extended the validity of a number of theories to new areas.

Perrin's concordance argument was simple and effective: by giving an impressive list of theories and experiments, by himself or others, that all led to comparable values for N, he made atoms the meaningful link between all these unrelated phenomena. Simultaneously his measurements supplied a relatively precise measurement of the number N and of the size of the molecules (or particles) in his experimental set-ups.

4. The ultramicroscope

The decisive experimental resource used by Perrin to connect the molecular and the macroscopic domains was the colloid solution. His experiments relied in large part on a series of recent investigations that had begun in 1902 with the elaboration of a new instrument, the ultramicroscope. This particular form of dark field microscope was devised in the Zeiss instrument making workshops in Jena by the colloid chemist Richard Zsigmondy and the Zeiss optician Henry Siedentopf (see Fig. 2).[37] It enabled the visualisation,

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[32] This was one of the most arduous tasks of the experiment, that earned Perrin much praise. Gouy himself wrote to Perrin in 1909 that 'if it had not been for the difficulty which I perceived in preparing such granules, I would have more actively pursued my experimentation on Brownian motion' (quoted in Nye, 1972, p. 105).

[33] To determine the radius of the grains Perrin: 1. used Stokes's Law; 2. counted the number of grains in a sample of the emulsion (and thus obtained their mass and radius, once their density was known), and 3. measured the radius directly by letting the emulsion dry: the grains then tended to adhere in lines that could be measured under the microscope.

[34] Perrin (1909), pp. 58–62.

[35] Einstein (1905), p. 549.

[36] Perrin (1913).

[37] On the elaboration of the ultramicrosope in the Zeisswerke, see Cahan (1996).

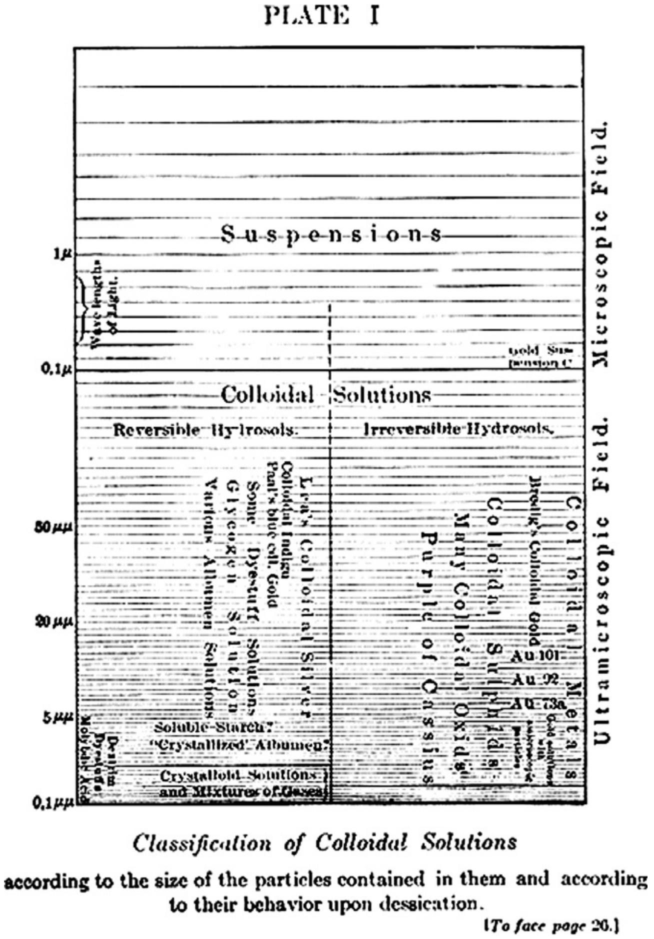

Fig. 3. Zsigmondy (1909), Plate 1.

or *Sichtbarmachung*, as its inceptors called it, of particles up to 500 times smaller than the smallest particles shown by an ordinary microscope. The ultramicroscope was able to overcome theoretical resolution (the maximum resolving power predicted by optical theory) though the *lateral* illumination of the particles against a dark background. Normally invisible particles were thereby revealed by the (much larger) cones of diffraction they produced under such conditions of lighting. According to the two scientists, and subsequent users, they appeared as bright spots of light on a dark background, similar to stars in the sky. As one early commentator put it, 'with this apparatus . . . measurements are possible that are not far from the so-called molecular dimensions, 0.1–0.6 μμ' representing an 'exceptional extension of the limits of sensory perception'.[38]

The ultramicroscope transformed colloid chemistry. For decades chemists had been puzzled by the composition of these liquids that behaved at times as solutions and at times as suspensions or emulsions, and that were refractory to the usual chemical tools of investigation, such as evaporation or precipitation. Zsigmondy, who had worked several years on colloid ceramic and glass pigments as an advisor to ceramic factories in Graz, imagined that the characteristic opalescence of colloid solutions was due to the presence of minute particles that diffracted light. The ultramicroscope enabled him to demonstrate that this was the case, and he identified a great number of colloid solutions as suspensions of ultramicroscopic particles, which behaved like solutions or like suspensions depending on the size of the particles.[39] Thereby Zsigmondy was able to establish a *continuum* between molecular solutions and the coarsest suspensions, with colloid solutions as intermediaries (see Fig. 3).[40]

After the publication of the results of Siedentopf and Zsigmondy's collaboration in the *Annalen der Physik* in 1903,[41] the ultramicroscope immediately attracted attention in all corners of the scientific world. Reviews and researches inspired by their work appeared in journals of physiology, chemistry, and physics all over Europe in the following months and years.[42] But the French were perhaps the quickest to take up ultramicroscopy outside of Germany. Aimé Cotton, physics lecturer at the Ecole Normale in Paris and Henri Mouton, a former Ecole Normale student now working at the Pasteur Institute, not only reviewed Siedentopf and Zsigmondy's work weeks after it was published, but they also proposed a modified ultramicroscopic arrangement as early as June 1903.[43] They wrote together the first textbook of ultramicroscopy (published 1906), where they detailed the technical set up as well as possible fields of application of the instrument.[44] It was probably from them that Jean Perrin learned about the new instrument.[45] Perrin began working on the contact electrisation of colloid particles in 1903, and in 1904 he acknowledged the ultramicroscope for the first time in print—the Siedentopf/Zsigmondy as well as the Cotton/Mouton types.[46] Cotton's laboratory notebooks of 1904–1905 testify to the proximity of his scientific interests to Perrin and Langevin's. The notebooks feature several passages concerning the motion of charged particles, and Cotton's attempts to measure, as Langevin had done with ions, their mean free path with the ratio of charge to mass e/m. If Cotton mentioned observations of the Brownian motion of colloid particles, he does not seem however to have dedicated any in-depth investigations to it, focussing instead on the electrical properties of colloid particles.[47]

When Perrin took up his study of Brownian motion in 1905 he relied crucially on the ultramicroscope and the recent results he and others had obtained with it in the investigation of colloid solutions. It was this instrument that made it possible for him to go beyond Gouy, who had regretted that the limits of optical resolution 'opposed an impassable obstacle to further progress, and we must

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[38] Biltz (1905), p. 300.

[39] Zsigmondy (1909).

[40] For this discovery Zsigmondy was awarded the chemistry Nobel prize in 1925 and a professorship for inorganic chemistry at the University of Göttingen; while Siedentopf became director of the Zeiss microscopy laboratory in 1907. *Carl Zeiss Jahresbericht* 1945/1946 Abt. Mikro (Historischer Teil), typescript in the Carl Zeiss Archiv, Jena.

[41] Siedentopf & Zsigmondy (1903b). The paper was accepted for publication in October 1902.

[42] For instance, Zsigmondy was invited to speak to the Royal Microscopical Society in London in June 1903; A Prof. Ernst discussed the ultramicroscope at the 78th Versammlung des ärtzlichen Centralvereins in Zurich in 1905 (LXVIII. Versammlung des ärtzllchen Centralvereins, 1905); articles on the instrument appeared internationally in a variety of journals, for example, Michaelis (1905), Puccianti (1904), Abbe (1904).

[43] Cotton & Mouton (1903) write that their apparatus was presented to the Académie des Sciences on 22 June 1903 and to the Société Française de Physique on 3 July 1903.

[44] Cotton & Mouton (1906).

[45] Perrin acknowledged the help of Henri Mouton in his first article mentioning the ultramicroscope: Perrin (1904–1905), p. 651. He had been on a familiar basis with both men since the late 1890s, as their correspondance testifies. See, for example Perrin's note announcing his nomination at the Sorbonne in a letter from Mouton to Cotton of 24 March 1898, Box 6, Fonds Aimé Cotton, Bibliothèque de Physique, Ecole Normale Supérieure, Paris.

[46] Perrin (1904–1905).

[47] Aimé Cotton's laboratory notebook entitled '1904–5 Ultramic', Box 13, Fonds Aimé Cotton, Bibliothèque de Physique, Ecole Normale Supérieure, Paris.

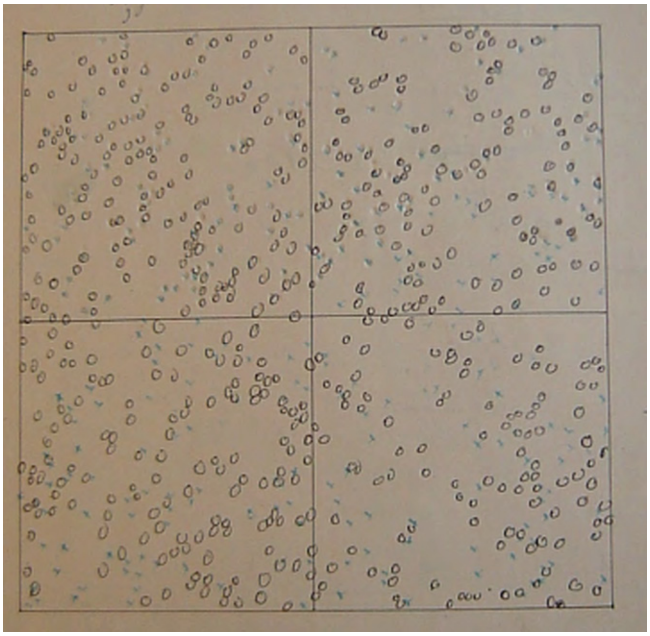

Fig. 4. Counting the number of particles in a sample using a microscope or ultramicroscope and a camera lucida. Jean Perrin, laboratory notebook (unpaginated, undated, ca. 1908). © Archives de l'Académie des Sciences, Paris.

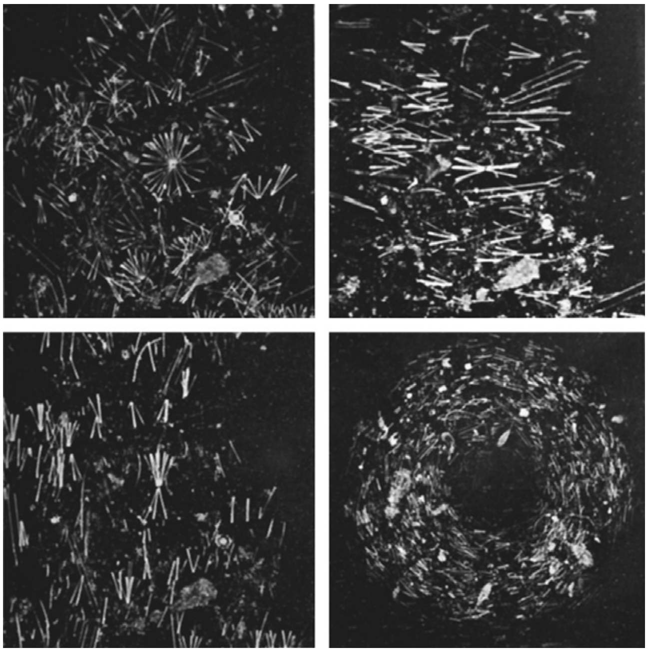

Fig. 5. 'Abhängigkeit der Sichtbarmachung vom Azimut der Beleuchtung' ('Dependence of visibility from the azimuth of illumination'), Siedentopf (1908), Plate IV.

give up the hope of one day seeing the phenomena and the beings concealed to our eyes by their small size'.[48]

Following the colloid researchers, Perrin painstakingly produced a colloid solution of gamboge, measured the density of gamboge and through observation of the colloid particles using the ultramicroscope he determined the number of particles in a given sample of solution (see Fig. 4). Perrin also found in the work of Siedentopf and Zsigmondy the experimental equivalent of his own theoretical mediation between the molecular and the macroscopic levels. The ultramicroscope revealed an intermediary dimension in which minute particles, whose size approached molecular dimensions, were visible and could be experimented upon. The behaviour of the particles made visible by the ultramicroscope supplied precious information on the size and the properties of molecules. Perrin relied on this argumentation in order to turn the metaphysical entity of the atom into a real, nearly graspable object.

The images produced by the ultramicroscope can however hardly be considered unmediated visual representations, for the ultramicroscope made neither atoms nor molecules nor even the much larger colloid particles visible, but only their diffraction cones; the particles were only made visible indirectly at the cost of showing their colour, their size and thickness.[49] An approximate value for their size could only be obtained through calculation taking into account the optical theory of the instrument and the laws of diffraction. Fig. 5, from an article by Siedentopf on ultramicroscopic technique, shows the dramatic changes in the appearance of the same solution depending on the angle of illumination, and hence the importance of training for producing reliable, reproducible evidence.

One way of building trust in his ultramicroscopic measurements was to carry out similar experiments using much larger particles visible in conventional microscopes. By using different experimental probes, Perrin was able to establish a continuum between his smallest and his largest suspended particles. We have here an experimental concordance through different means of visualization, a common form of argumentation especially in microscopic work. As Ian Hacking put it in *Representing and intervening*: 'we are convinced because instruments using entirely different physical principles lead us to observe pretty much the same structures in the same specimen'.[50] If suspensions of large particles observed using conventional microscopes behaved exactly as suspensions of the smallest particles observed under the ultramicroscope, then, Perrin argued, could this model not be applied to molecules in a solution, and conclude that molecules were discrete entities? He argued that

> This eternal motion ... is an already visible consequence of the regular molecular collisions against the particles ... the suspended grains function like visible molecules of a perfect gas.[51]

Perrin's argument rested on the theoretical and experimental concordance of values obtained using different methods; but also on establishing the continuity between the molecular and the macroscopic domains; such that he could claim that 'the observation of emulsions supplies an experimental basis for molecular theories'.[52]

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[48] Gouy (1895), p. 3. Aimé Cotton and Henri Mouton suggest that already in the 1880s Gouy developed optical methods similar to Siedentopf and Zsigmondy's (Cotton & Mouton, 1903, p. 1186).

[49] Siedentopf and Zsigmondy write: 'Die nicht unbeträchtlichen Erfolge bei der Untersuchung von Rubingläsern könnten aber leicht zu einer überschätzung unserer Methode, die Sichtbarmachung ultramikroskopischer Teilchen betreffend, Verlanlassung geben, der wir an dieser Stelle vorbeugen möchten. Vor allem sei hier hervorgehoben, dass unser Verfahren keinerlei Aufschluss über Form und Gestalt der kleinen Teilchen gibt; sie mögen wie auch immer geformt sein, stets wird man nur ein kleines Scheibchen als Beugungsbild erhalten. Nur wenn ultramikroskopische Teilchen so ausgebildet sind, dass eine ihrer Dimensionen grösser als eine halbe Wellenlänge wird, können sie unter dem Mikroskope als Stäbchen, Fäden oder elliptische Scheiben sichtbar werden. Die runden Beugungscheiben verschiedenartiger Teilchen besitzen aber je nach Grösse und Färbung derselben grosse Mannigfaltigkeiten der Helligkeit und Farbe' (Siedentopf & Zsigmondy 1903a, p. 11).

[50] Hacking (1983), p. 209.

[51] Perrin (1908), p. 967.

[52] Perrin (1909), pp. 111–113.

This last strategy matches perfectly the argument Perrin had put forward in *Les principes*: to consider atoms as belonging to the realm of *possible*, if not actual perception, and thereby to take them out of the realm of the metaphysical and into the laboratory, and treat molecules as Pasteur did microbes. Perrin's analogy with microbiology was incidentally literally realised when the ultramicroscope served to prove the existence of the bacteria causing syphilis. In 1905 Fritz Schaudinn and Erich Hoffmann had announced at the Berliner Medizinische Gesellschaft their discovery of an 'extraordinarily delicate, barely refracting, but vividly active spirochaete' responsible for syphilis.[53] Because it enabled the visualisation of the practically transparent spirochaete, and this without killing it through staining or heating, the ultramicroscope soon became an essential tool in the investigation of the spirochaete life-cyle and the transmission and evolution of the disease.[54] A comparison of colloid chemists', physicists and bacteriologists' take up of the ultramicroscope in the study of sub-microscopic entities provides enlightening clues on constrasting modalities of bridging the dimensions in different disciplines.

Perrin's use of the ultramicroscope was part of his efforts to promote physical chemistry. From early on, Perrin embraced the technologies and methods of physics to investigate chemical phenomena, as his doctoral work on cathode rays shows, or his enthusiasm for Röntgen's discovery of X-rays, leading him to publish only a few weeks later the results of his own experiments with these.[55] Like the other technologies Perrin employed, the ultramicroscope was made to provide evidence in support of the particulate view of matter, thereby showing matter to be amenable to study using the laws of mechanics (kinetic theory), and in turn validating the physical-chemical approach characteristic of his work and that of his closest friends and allies, Pierre and Marie Curie, Paul Langevin, and beyond, the Cavendish Laboratory scientists.

5. Seeing right

In the third section of his 1909 paper, Perrin attempted not only to determine the total energy of his emulsions, but also the molecular energy of individual colloid particles. This part brings out particularly clearly an aspect of Perrin's argument, that Brownian motion could only become an experimental, visual proof of atoms if it was observed *in the right manner*; and this involved a realisation that the sub-microscopic world followed rules of its own. Prior to his own work, Perrin maintained, the real meaning of Brownian motion had not be recognised; just as, no one, before Siedentopf and Zsigmondy, has recognised that colloid solutions were actually suspensions of minute particles. 'One thought wrongly' wrote Perrin, 'and believed that the shape of the grains could solve the question of heterogeneity'. 'One observed wrongly', he continued, referring to the ultramicroscope 'in that scientists placed themselves directly in the axis of the beam of sunlight, and were completely blinded'.[56]

Perrin first showed that earlier measurements were wrong: direct measurement of the velocity of individual particles was impossible due to the exceptionally rapid and variable motion of the particles. The values obtained until then were much too small (by a factor of 100 000), compared to the values predicted by the kinetic theory.[57] In order to obtain a meaningful measurement, Perrin used the formulae and methods developed by Albert Einstein for the determination of the average molecular energy of individual particles. Einstein had proposed to make displacement rather than instantaneous velocity the primary observable quantity, that is, the displacement of a particle during a given interval of time. Each displacement was measured by identifying the position of a particle at the beginning and at the end of this period, without taking into account all the intervening variations in velocity and direction. Perrin projected his emulsion using a camera lucida and followed the motions of individual particles; every thirty seconds he marked the position of the particle on a sheet of squared paper; subsequently he also used photography (see Fig. 6). The length of each section was then measured and squared. Avogadro's number could be obtained by calculating the average of half this value.

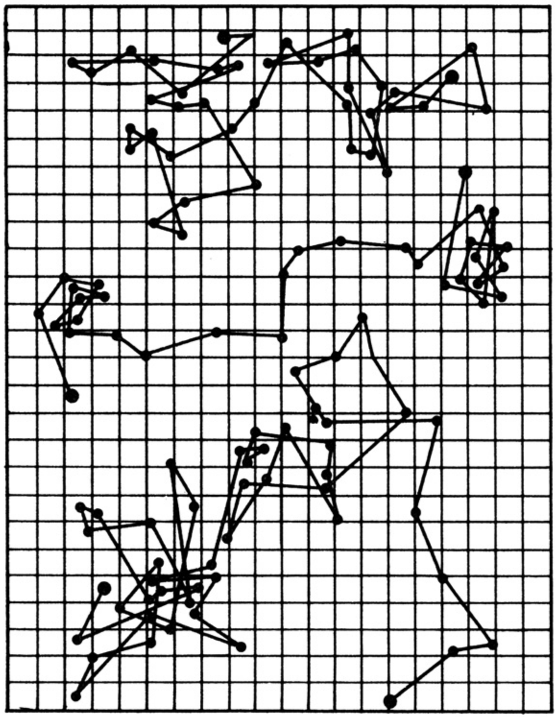

Fig. 6. Displacement of three particles: using a camera lucida Perrin marked the successive positions of each particle at regular intervals of time, before drawing straight lines to join the dots (Perrin, 1909, p. 81).

In doing so, Perrin critized other experimental attempts to verify Einstein's formula. The Svedberg had attempted to measure the motion of grains using the ultramicroscope, by moving a solution such that the luminescent trajectories of individual grains could be followed (using retinal persistence). Though Svedberg claimed to have confirmed Einstein's theories, Perrin claimed that Svedberg, 'had clearly been the victim of an illusion'.[58] Victor Henri's attempt using ultramicroscopy and cinematography to measure displacements were theoretically sound, he continued, but Henri's values were not trustworthy due to the latter's erroneous estimation of grain diameters.

Perrin's argument thus relied on a reconfiguration of the observation of Brownian motion. Even though the phenomenon had long been known and was easy to produce, it had not revealed its secrets, because no one had observed it in the right manner, using the right instruments and with the right theoretical apparatus. In

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[53] 'außerordentlich zarten, schwach lichtbrechenden, dabei aber sehr lebhaft beweglichen Spirochaeten' (Schaudinn & Hoffmann, 1905, p. 527).

[54] See for instance Lévaditi & Roché (1909).

[55] Perrin (1896), p. 186.

[56] Perrin (1904–1905).

[57] Perrin (1909), pp. 29–31.

[58] Perrin (1909), pp. 70–75.

the end, Perrin transformed a widely available phenomenon into evidence:

> In short, the mere observation of Brownian motion suffices to suggest that all liquids consist of elastic molecules in eternal motion.[59]

6. Visuality and atomic reality

Just as Maxwell's law was universally associated with its characteristic splattering of dots on a target, Perrin's diagram tracing the displacement of individual particles on squared paper became, and has remained, iconic of this new way of observing and of measuring Brownian motion. As his friend, the mathematician Emile Borel put it in 1912, in the observation of invidual atoms and corpuscules, e.g. in the study of the emission of alpha particles 'it is thus only this average number which exists scientifically'.[60] The broken lines of the curve may well represent the particular trajectory of a selected particle, whose position was marked at regular intervals of time. But this curve, or any other curve produced with different particles, different solutions, different time intervals stood for its underlying statistical stability: all measurements led to N. The curve stood for the realisation that the molecular dimension functioned differently from the macroscopic, but also that the former could be apprenhed using new methods. For mathematicians such as Borel, it was this numerical coincidence which certified atomic reality. Likewise Emile Picard wrote to Perrin in 1909: 'If things exist when numbers can be made to correspond to them, the existence of molecules can no longer be doubted'.[61]

But other scientists were more struck by the visual aspects of Perrin's argumentation.[62] The advocates of atomism in particular welcomed Perrin's experiments as constituting the first visible proof of atoms, or at the very least of their motion. Walther Nernst wrote in the 1909 edition of his textbook of physical chemistry that

> in view of the *ocular* [Augenfällig] confirmation of the picture which the kinetic theory provides us of the world of molecules, one must admit that this theory begins to lose its hypothetical character.[63]

A revealing description of Perrin's work by the Swedish mathematician Gösta Mittag-Leffler goes in the same direction. In a letter to Poincaré in 1909 he wrote that 'it consists of a method of isolating and seeing atoms'.[64] More accurately, but no less vividly, Rutherford told the mathematical and physical section of the BAAS in 1909 that:

> The character of the Brownian movement irresistibly impresses the observer with the idea that the particles are hurled hither and tither by the action of forces resident in the solution, and that these can only arise from the continuous and ceaseless movement of the invisible molecules of which the fluid is composed.[65]

William Crookes, himself the inventor a fews years earlier of the spinthariscope, a device for visualizing what he believed to be 'positive atoms' individually, was delighted.[66] In October 1910, he wrote to Perrin inviting him to give a lecture at the Royal Institution as one of the 'scientific men of great eminence'.[67] Wolfgang Ostwald, colloid chemist and son of energeticist Wilhelm Ostwald wrote in the 1909 edition of his *Grundriss der Kolloidchemie* that

> The isolation and enumeration of gas ions on the one hand, which have crowned with full success the long and remarkable labours of J. J. Thomson, and on the other hand the agreement of the Brownian motion with the requirements of the kinetic hypothesis carried out by a series of researches, most lately and completely Jean Perrin, now justify even the cautious scientist to speak of an experimental proof of the atomic constitution of the space-filling matter.[68]

Though the ultramicroscope did not actually make atoms visible, its visualizing power seems nonetheless to have been decisive in the general perception that atoms were concrete and therefore real objects. In providing conclusive evidence of the reality of atoms, the ultramicroscope therefore played a symbolic as well as a practical role. In order to make his argument, Perrin further relied on an implicit analogy between colloid particles and molecules; he also made use of a body of theories (kinetic theory of gases applied to liquids, Stokes's law modified to apply to small particles, diffusion theory, and so on) in order to turn the Brownian motion of colloids into evidence of atomic reality. Perrin claimed that his abilities as theoretician and observer enabled him to properly understand and observe Brownian motion. In this way Perrin was able to drag the atomic debates from the philosophical realm and into the experimental one, and to bring visual evidence without actually showing images of atoms or molecules.

Perrin's optimism about the possibility of visualizing, albeit indirectly, the sub-microscopic world was shared by many contemporaries, as the reception of his work by physicists and chemists show. Rutherford, summarizing his view on the contribution of recent research in the atomic realm in his 1909 speech to the British Association for the Advancement of Science claimed:

> I should like to emphasize the simplicity and directness of the methods of attack on atomic problems opened up by recent discoveries. As we have seen, not only is it a simple matter, for example, to count the number of particles by the scintillations produced on a zinc sulphide screen, but it is possible to examine directly the deflection of an individual particle in passing through a magnetic or electric field, and to determine the deviation of each particle from a rectilinear path due to encounters with molecules of matter. We can determine directly the mass of each particle, its charge, and its velocity, and can deduce at once the number of atoms present in a given weight of any known kind of matter. In the light of these and similar direct deductions, based on a minimum amount of assumption, the physicists have, I think, some justification for their faith that

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[59] Perrin (1909), pp. 12–15.

[60] Borel (1912), p. 847.

[61] Emile Picard to Jean Perrin, 30 November 1909, Fonds Jean Perrin 54J, Arch. Acad. Sci., Paris.

[62] This is not to suggest that Perrin's work was not criticised. The 1809 *Annales* paper was the last and most comprehensive of a series of papers by Perrin, which integrated responses to various comments and criticisms made on earlier work. For a discussion of critiques of Perrin's work in these years (mostly by Jacques Duclaux, Victor Henri and Aimé Cotton) and of other experimental work inspired by Einstein's paper, see Nye (1972), pp. 97–143.

[63] Nernst (1909), p. 212, translated by Brush (1968), p. 35.

[64] Gösta Mittag-Leffler to Henri Poincaré, 13 March 1909, Archives Narbonnaud (1999), p. 243.

[65] Later on he added: this 'is thus a striking if somewhat indirect proof of the general correctness of the kinetic theory of matter' (Rutherford, 1909, p. 292). Perrin's paper was translated by F. Soddy and published in 1910; many scientists, though clearly not Rutherford, became aware of Perrin's work only then.

[66] Crookes (1903), p. 241.

[67] William Crookes to Jean Perrin, 24 October 1910, Fonds Jean Perrin 54 J, Arch. Acad. Sci., Paris.

[68] Wo. Ostwald, introduction to what was to become (Ostwald, 1909): single page enclosed in a letter to Jean Perrin dated 14 October 1909, Fonds Perrin 54J, Arch. Acad. Sci., Paris.

> they are building on the solid rock of fact, and not, as we are often so solemnly warned by some of our scientific brethen, on the shifting sands of imaginative hypothesis.[69]

The major difference between, say J. J. Thomson's work on the electron and Perrin's work was that the latter explicitely framed his investigations as a solution to the philosophical, epistemological problem of atomism rather than as the discovery of a new effect or sub-microscopic particle. Perrin's concern for the larger questions in physics and chemistry, his ambition to found a French field of physical chemistry led him to present his work on colloid particles and Brownian motion as the solution to a major scientific debate of the nineteenth century. Yet Perrin and his contemporaries' optimistic belief in the commensurability of the molecular, atomic and subatomic dimensions with the macroscopic world, occupy a singular, ultimately short-lived place in the history of the physical sciences.[70] The new sense of reality which physical scientists might have developed in the early years of the twentieth century was rapidly overhauled, beginning with Niels Bohr's quantum model of the atom of 1913. Perrin himself, who launched in the 1910s a series of experiments on thin layers aimed at visualizing the superposition of molecules in layers of liquid, found it increasingly difficult to uphold his earlier views. Towards the end of his life he admitted that

> we used to think that it would be possible ... to study nature in portions of decreasing size, thanks to a magnification without limits ... But we have since understood that all concepts, even that of Space, was formed at a particular scale of observations, and must cease to be valid for conditions diverging infinitely from those in which it was built.[71]

7. Conclusion

At the beginning of his long article of 1908, in which his Brownian motion investigations were brought together, Perrin explained in much detail how to project Brownian motion for a large public, suggesting that he began his public lectures with such a demonstration of this phenomenon. Subsequently he had films on atoms made for him by the pioneer of scientific cinematography (and ultramicro-cinematography) Jean Comandon. Perrin's films were repeatedly showed at public conferences.[72] The care taken by Perrin in producing these visual displays of Brownian motion for his publics is symptomatic of a pervasive concern for visuality that was expressed in his own epistemology, research and conception of scientific research. The beauty of Perrin's demonstration was that he was able, before the eyes of his public, to transform the meaning of Brownian motion. At the end of his presentation, this projected random movements of the particles had become evidence of atomic reality, and its icon the displacement diagram. It is perhaps revealing that in the interwar as he increasingly moved away from laboratory work and into politics and science policy, one of Perrin's major projects was nonetheless centrally concerned with visualization: the creation in 1937 of the first large science museum in France, the Palais de la Découverte, in which 'living science' was to be presented to the public, and that included a section in which Perrin's own Brownian motion experiments could be visualized.

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[69] Rutherford (1909), p. 302.

[70] For a comparative perspective on the Brownian motion experiments and sense of visuality of Perrin's contemporaries The Svedberg and Max Seddig, see Lundgren (1993) and Scott Curtis (2005) respectively.

[71] Perrin (1948), pp. 69–70.

[72] The earliest reference found to such a film is 1911: Perrin (1911), p. 776. Perrin repeatedly ordered Brownian motion films from Comandon as shown by his collaborator André Marcelin's letter to Comandon in 1921, Fonds Comandon, Box COM B1, Archives de l'Institut Pasteur, Paris.

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