# Controlling the Invisible: Experimental Strategies and Hypotheses in Discovering the Cause of Brownian Movement

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## 1. Introduction

Brownian movement is the seemingly completely irregular movement of microscopic particles—of a diameter less than approximately 10-3 mm-of solid matter when suspended in liquids.<sup>1</sup> Although it was experimentally investigated throughout the nineteenth century, it was only at the end of that century that the importance of this phenomenon for the kinetic-molecular theory of matter-i.e., the theory that matter is composed of atoms and molecules in incessant motion-was finally recognized. Historians of science have expressed surprise and lament about the fact that Brownian movement played no role in the early development and justification of the kinetic theory of gases. Today we know that Brownian movement is a directly observable effect of the motion of molecules constituting the liquid state of matter. If molecular motion had been identified from the beginning as the cause of the phenomenon, some of the most important philosophical and scientific objections raised against the early kinetic theory could have been answered. For example, the molecular explanation of Brownian movement could have resolved the nineteenth century philosophical debates over the empirical status of molecular hypotheses, which centered on the question of whether the existence of unobservable entities such as atoms and molecules could be resolved by means of observation and experiment. In addition, Brownian movement could have been used to provide independent empirical evidence for one controversial claim made in the kinetic theory of gases; namely that at a molecular level, the Second Law of Thermodynamics had only statistical, as opposed to absolute, validity. Related to this historiographical position we often find the claim that most of the nineteenth century experimental investigations on the nature and cause(s) of Brownian movement were somewhat less rigorous compared to later, early twentieth century experiments that successfully established molecular motion as the proper and unique cause of the phenomenon (Brush 1968, 1; Nye 1972, 9; Maiocchi 1990).<sup>2</sup>

In this chapter we focus on the experimental practices and the reasoning strategies followed by nineteenth century investigators of Brownian movement in their quest to determine the causal origin of the phenomenon. By focusing on these experimental practices and reasoning strategies, we gain a better appreciation of the nineteenth century investigative efforts in and of themselves, as opposed to insofar as they relate to later scientific and methodological developments. Nevertheless, our account present some of the practical and conceptual complexities of the investigations on the cause of Brownian movement that help to make sense of its delayed connection with the kinetic-molecular theory of matter. We argue that there was an extensive and sophisticated experimental work done on the phenomenon of Brownian movement throughout the nineteenth century. Most investigators were fully aware of the methodological strategies that they employed.

The first was the traditional strategy of varying the experimental parameters to discover causal relations. In the nineteenth century, this strategy was codified into explicit methodological rules by John Herschel (1830) and then, perhaps more famously, by John Stuart Mill ([1843] 1974). In the nineteenth century investigations of Brownian movement, we find that the reasoning underlying this strategy was already embedded in experimental practices before, and independently of Herschel and Mill (see also the

<sup>&</sup>lt;sup>1</sup> Brownian movement, mouvement Brownien, moto Browniano, Molecularbewegungen were the terms used in the nineteenth century to refer to the movement of microscopic particles suspended in liquids. In this chapter we use these terms to describe the nineteenth century investigations of this phenomenon. We avoid the term *Brownian motion*, which is more recent, already encompasses the randomness of the motions, and has wider connotations. According to Encyclopedia Britannica, for example, 'Brownian motion' concerns "various physical phenomena in which some quantity is constantly undergoing small, random fluctuations" (Britannica, 21 Mar. 2023).

<sup>&</sup>lt;sup>2</sup> These sentiments echo those of the historical actors who played a protagonist role in establishing the kineticmolecular explanation of Brownian movement. See, for example, Perrin 1910, and Poincaré 1905.

chapters by Schürch and Nickelsen, this volume). More specifically, the basic rationale underlying these investigations was that: (a) all the circumstances and factors which could be varied or entirely excluded without influencing Brownian movement, were not causes of the phenomenon, (b) all the circumstances and factors whose exclusion or variation influenced the phenomenon, were considered to play a causal role in its production. As mentioned in the introduction to this volume, the employment of this strategy required (implicitly or explicitly) at least three notions of control: (a) control over the circumstances or factor whose exclusion or variation was to be examined, (b) control over the rest of the circumstances or factors, which ought to be kept as much as possible the same, and (c) control in the more familiar sense of comparing the experimental situation with or after the intervention (i.e., the variation or exclusion of the factor whose causal influence was being investigated) with the (control) situation without or before the intervention (see also Boring 1954 and Schickore 2019).

The strategy of varying the circumstances succeeded more in excluding various suspected causal factors than in establishing a positive causal explanation of the phenomenon. Even when some causal influence was detected, the conclusion was not shared by all investigators. The existence of disagreements about the influence of various causal factors led to the recognition of the importance of a different notion of 'control': that of the independent confirmation of experimental results by other researchers. Despite the difficulties surrounding its implementation, the strategy of varying the circumstances, by showing the insufficiency of the various causal explanations of Brownian movement, increased the importance of the fact that the newly developed kinetic-molecular conception of matter provided a plausible explanation of the phenomenon.

The second strategy was similar to—what at the time was called—the *method of the hypothesis*. The latter, at least according to some scholars, re-emerged in the nineteenth century as the proper strategy for validating explanatory hypotheses about unobservable entities, processes, and phenomena (Laudan 1981). Amidst all the criteria for evaluating explanatory hypotheses, the ability of a hypothesis to explain, successfully predict, and/or be supported by a variety of facts—especially facts that played no role in the hypothesis' initial formulation—was considered to be the most important criterion indicating its validity. Proponents of this strategy appealed to the ability of the kinetic-molecular hypothesis to offer a natural explanation of Brownian movement. What was remarkable about this explanation, they argued, was the fact that the elements of the hypothesis invoked to explain the phenomenon were developed independently of it. The ability of the kinetic-molecular hypothesis to explain a variety of unrelated phenomena and experimental evidence was offered, by some investigators, as an important way to 'control' the validity of the kinetic-molecular explanation of Brownian movement.

None of these two methodological strategies could, on its own, establish molecular motion as the cause of Brownian movement. The combination of these two strategies and their accompanying notions and practices of control, led, at the end of the nineteenth century, to the recognition of molecular motion as the most probable cause of the phenomenon. From then on, the goal of experimental practices and reasoning strategies shifted to that of probing and evaluating the kinetic-molecular explanation of Brownian motion.

## 2. First Observations of the Curious Phenomenon

The phenomenon of Brownian movement owes its name to the Scottish botanist Robert Brown (1773-1858) who experimentally investigated it, beginning in the summer of 1827 (Brown 1828). An already eminent botanist at the time, Brown was not the first to observe the phenomenon. All earlier investigators, however, seem to have connected it with the motion of infusory animalculæ, and had attributed it to some sort of vitality possessed by the moving particles (Brown 1829, 164; Brush 1968). Brown's main contribution, and his claim to priority lies in establishing that the movement of microscopic particles when suspended in liquids was a general phenomenon exhibited by all microscopic particles, independently of their chemical nature. We start by examining the methodological ideas and practices that Brown used to establish this claim.

Brown offered an account of his initial investigations in a pamphlet he originally circulated privately among his friends, but which aroused enough interest to appear, in 1828, in the *Edinburgh New Philosophical Journal*, and soon afterwards in numerous other journals (Mabberley 1985). The pamphlet provides an interesting step by step account of his investigations. Brown was investigating the mechanism of fertilization in the plant *Clarckia pulchella*, whose grains of pollen were filled with microscopic particles

of different size that were easy to observe with a simple microscope. "While examining the form of these particles immersed in water, I observed many of them evidently in motion; ... These motions were such as to satisfy me, after frequently repeated observations that *they arose neither from currents in the fluid, nor from its gradual evaporation, but belonged to the particle itself* (Brown 1828, 162-63, italics added).

Brown extended his observations to particles derived from the pollen of plants belonging to different families, and he invariantly found similar particles exhibiting the same spontaneous movements when suspended in water. Having found these movements in the particles of pollen of all the living plants that he examined, Brown inquired whether they continued after the death of the plant, and for how long they were retained (Brown 1828, 164). Unexpectedly, he found that specimens of dead plants, some of which were preserved in an herbarium for no less than one hundred years produced similar moving particles. Soon he discovered that the moving particles—or *active molecules*, as he began to call the smallest particles of apparently spherical shape not exceeding in size the 1/15000 of an inch—where not limited to the grains of pollen, but they could be produced from other parts of the plant as well. Even more surprisingly, however, Brown found that these molecules were not limited to organic matter but could be equally acquired from inorganic matter. He found that fragments of window glass, various minerals,

[r]ocks of all ages, including those in which organic remains have never been found, yielded the molecules in abundance. Their existence was ascertained in each of the constituent minerals of granite, a fragment of the Sphinx being one of the specimens examined...In a word, in every mineral which I could reduce to a powder, sufficiently fine to be temporarily suspended in water, I found these molecules more or less copiously (Brown 1828, 167).

The next step for Brown was to investigate whether the movement of the molecules derived from organic substances was affected by the application of intense heat on the substance from which they were derived. A comparative experiment was conducted. Small portions of wood (both living and dead), linen, paper, cotton, wool, silk, and hair were heated, and immediately quenched in water. In all cases, molecules could be derived, and they were found to be as evidently in motion as those obtained from the same substances before burning (Brown 1828, 168).

To sum up, during these initial investigations, Brown used the seeming invariance of the suspended particles' motion to the variation or change of the suspected causal factors—namely, currents and evaporation in the suspending liquid, chemical nature of the suspended particles, application of heat on the particles' originating material—to conclude the causal independence of these motions from the varied factors.<sup>3</sup> As already mentioned, this strategy of varying the circumstances to discover causal dependencies involves at least three notions of control: (a) control over the variation of the suspected causal factor, (b) control over the rest of circumstances which should remain as much as possible the same,<sup>4</sup> and (c) control in the sense of comparing the experimental situation with or after the variation of the suspected causal factor with the situation without or before the variation. Brown did not use the term 'control', and these three notions of control are only implied in the description of his observations and experiments. In the rest of the chapter, we will see that these and other forms of control became more explicit when the validity of the initial observations of the phenomenon were challenged.

The invariance of the phenomenon to the variation of some of the suspected causal factors, led Brown to exclude these factors from being causes of the surprising phenomenon, but could of course not help him identify a positive cause. His conclusions regarding the cause of the motion of the 'active molecules' were cautious: "I shall not at present enter in any additional details, nor shall I hazard any conjecture whatever respecting these molecules, which appear to be of such general existence in organic as well as inorganic bodies" (Brown 1828, 169).

In the pamphlet presenting the results of his early research, Brown stated that he knew close to nothing about the phenomenon before he began his investigations, and that he was only acquainted with the abstract of a memoir that the French botanist Adolphe Brongniart (1801-1876) had read before *l'Académie des Sciences* in Paris, in December 1826, and later published in the *Annales des Sciences Naturelles* (Brown 1828, 171-72; Brongniart 1827). Brongniart was also studying the process of fertilization in plants.

<sup>&</sup>lt;sup>3</sup> This early use of the varying the circumstances strategy belongs to what Steinle (2002, 2016) has identified as *exploratory experimentation*.

<sup>&</sup>lt;sup>4</sup> Brown explicitly stated that to give greater consistency to his statements, and to bring the subject as much as possible to the reach of general observation, he continued to use the same microscope with one and the same lens, throughout his initial investigations (Brown 1828, 161).

Using an Amici microscope, which provided a magnification of up to 1050 times in diameter, Brongniart found that the microscopic granules contained in the pollen grains of numerous plants (or "granules spermatiques", as he called them)—which formed "la poussiere fecondant", i.e., the most essential part of the pollen fertilizing the ovum—when suspended in water performed clearly distinguishable spontaneous movements, which seemed impossible to be attributed to an external cause (Brongniart 1827, 45). These observations corroborated, according to Brongniart, his initial hypothesis that the spermatic granules found in the pollen of plants were the analogous of the *spermatic animalcula* found "swimming" in the sperm of animals (Brongniart 1827, 48).

Published in some of the most prestigious scientific journals of the time, Brown's and Brongniart's observations drew a lot of attention, and also elicited a strong reaction against the claim that the moving microscopic particles were self-animated.<sup>5</sup> The most influential critique came from the French physiologist François Raspail (1794-1878), who claimed his conclusions on the subject to be the result of many repeated and varied experiments (Raspail 1829a; 1829b). First, Raspail attacked Brongniart claim that the granules that are discharged in the explosion of grains of pollen were analogous to the spermatic animalcules. His numerous experiments, argued Raspail, clearly showed that the granules derived from the explosion of the grains of pollen, of even the same plant, varied in shape, diameter, size, as well as other characteristics (Raspail 1829a, 97). This challenged the claim that these granules were of an organized nature and/or that they belonged to a distinguishable category of entities. Second, Raspail rejected the claim that the movements of the particles suspended in water belonged to the particles themselves. He argued that the movements were easily distinguishable from the spontaneous movements of the infusory animalcules, and that they could be easily attributed to the influence of various mechanical causes (Raspail 1829a, 1829b). Raspail listed several such causes which, based on "a great number of consecutive observations" (Raspail 1829a, 97), could communicate even to the most inactive particles the appearance of spontaneous motion. The list included the motion communicated to the granules from the explosion of pollen that discharges them, capillarity, the evaporation of the suspending water, the evaporation of the volatile substances with which the granules issuing from pollen may be impregnated, the ordinary motions of great towns, the motions caused by the air's agitation, the motions caused by the observer's hands, the inclination of the object plate, and the electricity communicated to particles of metallic origin by friction (Raspail 1829a, 97; Raspail 1829b, 106-7).

Raspail's list proved to be very influential. For the most part of the nineteenth century, it constituted the essential list of causes which, singly or in combination, were invoked to explain the movements of microscopic solid particles suspended in liquids. The list is also important because it reveals the difficulties surrounding the ascertainment of the concrete cause(s) of the observed movements by following the experimental strategy of varying the circumstances. Such an experimental effort would require a meticulous and rigorous control over all the numerous suspected causes and possible confounding factors.<sup>6</sup> Regarding his own methodological efforts, Raspail maintained that, although his numerous earlier experiments on the subject had made him aware of all the various contributing causal factors, faced with the solemn claims about the existence of spontaneous motion, he felt it incumbent on himself "to repeat all my experiments, and to vary them in every way, as if I had doubted the accuracy of my former ones" (Raspail 1829a, 99).

Replying to the criticism, Brongniart defended his original observations on both methodological and experimental grounds. Besides claiming his conclusions to be the result of repeated experiments performed on pollen from different kinds of plants, Brongniart appealed to another kind of experimental control: that of independent confirmation by other researchers.<sup>7</sup> Independent confirmation, Brongniart asserted, was essential for the verification of claims concerning phenomena that were not readily observable and which contradicted in certain respects widely established theories.<sup>8</sup> Brongniart emphasized

<sup>&</sup>lt;sup>5</sup> Brush 1968 provides an extended bibliography of these reactions.

<sup>&</sup>lt;sup>6</sup> Schickore (2022) and Schürch (this volume) provide detailed accounts of the difficulties surrounding the concrete applications of the varying the circumstances strategy to establish causal claims.

<sup>&</sup>lt;sup>7</sup> This kind of experimental control is discussed in detail in the chapters by Schürch and Christopoulou and Arabatzis.

<sup>&</sup>lt;sup>8</sup> "Les phénomènes de la nature, qui s'éloignent de ceux qui frappent habituellement nos yeux, qui contredisent à quelques égards les systèmes fondés sur des observations anciennes et généralement reconnues; qui, par cette raison, sont d'ordinaire plus difficiles à saisir, exigent, pour être admis au nombre des vérités non contestées, des recherches souvent répétées, présentées avec ces détails qui éloignent toute espèce de doute, et vérifiées par de observateurs

especially the fact that some of this confirmation came from research that was done without prior knowledge of his conclusions (Brongniart 1828, 392-3).<sup>9</sup> This specific kind of independent confirmation was important because it precluded the possibility that the other researchers had simply adjusted their conclusions to achieve consensus.<sup>10</sup>Among the claims that Brongniart maintained to have been independently confirmed by other researchers were the claims that the granules contained in the pollen of the same plant were of a well determined form, had exactly measurable dimensions, and that each one of them performed extremely small motions which, because of their irregularities, seemed to be independent of any external cause (Brongniart 1828, 382). To these independently confirmed observations, Brongniart added new observations conducted on twenty-four species of plants from different families as well as new experiments which, he claimed, established without any doubt that the 'spermatic granules' were different from the irregularly shaped particles of non-organized matter also found in the pollen of plants (Brongniart 1828, 386-88).

Regarding the movement of the 'spermatic granules', Brongniart cited the irregular way they changed their positions relative to one another to argue that it was not caused by any external influences but was dependent on a cause existing in the granules themselves (Brongniart 1828, 389). He too used the strategy of varying the circumstances to show that the movement continued without presenting the smallest difference even when some of the mechanical causes in Raspail's list—namely the agitation of the liquid caused from evaporation, the trembling of ground or air, and the influence of sunlight—were either excluded or varied. More specifically, Brongniart burst the grains of pollen in very small glass capsules made using small concave lenses, and which were filled with a drop of water. He then covered the capsules using a very thin film of mica to avert the evaporation and the agitation of the surface of the suspected mechanical causes, the movements of this preparation under the lamp light but also during cloudy days. Despite the measures taken to control, i.e., to exclude or lessen the influence of the suspected mechanical causes, the movements of the suspended granules continued without presenting the smallest difference. On the contrast, when he replaced water with alcohol in the same experimental setting, the movements ceased completely instead of becoming livelier as one would expect if they were caused by the liquid's evaporation (Brongniart 1828, 389-90).

Of special interest is the note additionelle to the paper which Brongniart wrote after learning about Brown's observations of the irregular movement of suspended particles derived from inorganic matter (Brongniart 1828, 393-98). Brongniart stated that Brown's observations prompted him to conduct new observations on suspended inorganic particles which, in general, agreed with those of the Scottish botanist.<sup>11</sup> Given that Brongniart's initial observations and experiments claimed to have established that the 'spermatic granules' in pollen were analogues of the spermatic animalcules in the sperm of animals, and that they were clearly distinguishable regarding both their form and movement from the (irregularly shaped) microscopic agglomerations of matter also found in pollen, the assertion of agreement with Brown's observations was certainly an exaggeration. In fact, even in the note, Brongniart continued to draw a distinction between the movement of the 'spermatic granules' contained in pollen, from that of inorganic particles. The movements of the inorganic particles seemed to him to be less constant and dependent on the nature of the inorganic substance from which they were derived. In general, the movements were more evident in inorganic particles derived from substances that were better conductors of electricity. Despite the differences between his observations and Brown's, and in line with his previous assertion about the importance of independent confirmation, Brongniart was eager to emphasize the points of agreement. The most important point of agreement consisted in the claim that the movements of both the spermatic granules and inorganic particles seemed to be caused by a force inherent in the

différens; car le concours des opinions d'hommes indépendans les uns des autres, est la seule preuve de la vérité pour ceux qui ne peuvent pas la rechercher eux-mêmes" (Brongniart 1828, 381-82).

<sup>&</sup>lt;sup>9</sup> "Cette observation est d'autant plus curieuse qu'elle a été faite par une botaniste qui ne pouvait avoir à cette époque aucune connaissance des résultats auxquels l'examen du pollen des plants phanérogames m'avait amené; qui n'y était conduit par aucune théorie, et qui même, par ces raisons n'a pas pu sentir la liaison de ces phénomènes avec d'autres analogues" (Brongniart 1828, 393).

<sup>&</sup>lt;sup>10</sup> For a discussion of this notion of (genetically) independent confirmation and its differences from other notions of independent confirmation see Soler 2012 and Coko 2020b.

<sup>&</sup>lt;sup>11</sup> "Quant aux molécules des corps inorganiques, on observe en effet assez souvent, dans plusieurs substances broyées dans l'eau de très-petits corpuscules arrondis semblables aux plus petites molécules du pollen, et doués de mouvemens analogues en apparence à ceux des granules du pollen" (Brongniart 1828, 394).

particles and not by any external factors.<sup>12</sup> The crucial point, he continued, was to determine whether they were due to the same cause(s). Especially, determining whether they were caused by some sort of vitality possessed by the particles, or by some other hitherto unaccounted internal factor or external influence (Brongniart 1828, 394-96).

Brown too rejected the charge that in his original memoir he had implied that the moving suspended particles were animated (Brown 1829, 161-62). He too claimed to have conducted additional research on the subject, this time using different microscopes, and different kinds of particles suspended in various liquids (Brown 1829, 162). The additional research, Brown asserted, confirmed the main results he had advanced in his 1828 pamphlet, namely:

that extremely minute particles of matter, whether obtained from organic or inorganic substances, when suspended in pure water, or in some other aqueous fluids, exhibit motions for which I am unable to account, and which from their irregularity and seeming independence resemble in a remarkable degree the less rapid motions of some of the simplest animalcules of infusions...I have formerly stated my belief that these motions of the particles neither arose from currents in the fluid containing them, nor depended on that intestine motion which may be supposed to accompany its evaporation (Brown 1829, 162).

Brown cited the complete irregularity of the movements—i.e., the seemingly complete independence in the movements of every two particles—to reject the various mechanical explanations of the phenomenon. In addition, he described two experiments demonstrating that the particles continued to move with their usual degree of activity, while the principal mechanical causes suspected of their motion were either reduced or completely excluded.

In the first experiment, Brown was able to isolate minute drops of water, some of them containing few or only one microscopic particle, in almond oil. In this manner the minute drops of water, which if exposed to air would be dissipated in less than a minute, were retained for more than an hour. But in all drops of water thus formed and protected the motion of the suspended particles continued with undiminished activity, while the mechanical causes suspected of their motion, namely evaporation and the particles' mutual attractions and repulsions, were either reduced or entirely excluded.

In the second experiment, Brown was able to show that the motion of the particles was not produced by causes acting on the surface of the water drop—e.g., currents in the surrounding liquid. Inverting his first experiment, Brown mixed a very small proportion of almond oil with the water drops containing the particles and was able to produce microscopic almond oil drops of extreme minuteness, some of them not exceeding in size the particles themselves, attached to the surface of the water drops. The oil drops remained nearly or altogether at rest while the material particles isolated in the water drops continued to move with their usual degree of activity (Brown 1829, 163-64).

Brown and Brongniart's observations and experiments seemed to have to have aroused a lot of interest and debate regarding the cause of the curious phenomenon. Because many researchers considered vitalist explanations questionable at the time, the idea of the vitality of the particles was rejected and, despite Brown and Brongniart's experimental efforts, various mechanical causes, singly or in combination, were proposed as explanations. Georg Wilhelm Muncke from Heidelberg cited experimental research on the phenomenon, to conclude, in 1829, that: "The movement certainly bears some resemblance to the one observed in Infusoria, yet the latter shows more voluntary action. Vitality, like many possibly have believed, is out of the consideration [as an explanation]. I rather consider the motion to be purely mechanical and caused by the uneven temperatures in strongly illuminated water, evaporation, air and heat currents, etc."<sup>13</sup>

<sup>&</sup>lt;sup>12</sup> "La seule chose sur laquelle je ne puis conserver aucun doute, et sur laquelle j'ai le bonheur de voir mon opinion entièrement confirmée par celle des commissaires de l'Académie et de M. Brown, c'est l'indépendance complète de ce mouvement de toutes les causes extérieures influant sur le liquide ambiant. Il me paraît bien certain que la cause du mouvement, quelle quelle soit, réside dans une force physique ou organique inhérente aux corpuscules mêmes qui se mouvent. C'était la seule chose que j'avais avancée dans mes premières observations sur ce sujet, puisqu'en disant que ce mouvement était spontané, j'avais observé que j'entendais seulement exprimer par ce mot que ce mouvement était inhérent aux granules eux-mêmes" (Brongniart 1828, 396).

<sup>&</sup>lt;sup>13</sup> "Die Bewegung hat allerdings einige Aehnlichkeit mit der bei Infusorien wahrgenommenen, jedoch zeigt letztere mehr Willkühr. An Vitalität, wie vielleicht Einige geglaubt haben, ist dabei gar nicht zu denken, vielmehr halte ich

These mechanical explanations of the phenomenon persisted, despite Brown's and Brongniart's experiments showing the phenomenon's invariance even when explicit measures to control and/or exclude the influence of the relevant mechanical causes were taken. It seems that one important factor for the persistence of the various mechanical explanations was the impression that their rejection would leave the particles' vitality as the only plausible explanation. For example, the renowned Scottish physicist David Brewster, editor at the time of the *Edinburgh Journal of Science*, referring to the sufficiency of Raspail's mechanical causes to explain the motions of the suspended particles, remarked that "even if they did not afford a sufficient explanation of the motions in question;—nay, if these motions resisted every method of explanation, it is the last supposition in philosophy that they are owing to animal life" (Brewster 1829, 219). For Brewster, an explanation showing that the motions of the suspended particles obey physical laws like the ones governing the motions of larger bodies, would always take precedence over any hypothesis claiming the particles to be in some way animated (Brewster 1829, 219-20).

## 3. Experimental Investigations of Brownian Movement: 1830-1860

Despite the lack of agreement regarding the causal origin of the curious phenomenon, Brownian movement was not neglected during the period 1830-1860, as sometimes claimed. In fact, we can say that what was neglected were some of the investigators of the phenomenon by subsequent historiography of science. One of these neglected figures was Giuseppe Domenico Botto (1791-1865), professor of experimental physics at the University of Torino, who conducted experimental investigations on the phenomenon in the late 1830s (Guareschi 1913). Fully aware of the existing disagreements regarding the characteristics and causes of the phenomenon, Botto called for a cautious, purely experimental approach, and for a multiplication of experiments.<sup>14</sup>

In his own investigations, Botto found that the movement of suspended particles derived from organic matter had different characteristics from that of inorganic particles. Using an Amici horizontal microscope Botto conducted extensive microscopical observations on suspended microscopic globules derived from different plants, vegetable products, and inorganic substances. In all his observations of suspended microscopic globules derived from vegetable matter Botto found the phenomenon exhibited in the manner described by Brown: "one sees them changing their relative positions every moment, approaching one another, receding from one another, spinning, as if these movements originated on their own."15 The lively oscillatory movement was invariantly found on suspended globules derived from all the parts of the individual plant: the grains of pollen, the ovary before and after fertilization, the pistil, the stamen, the anther, the buds, the tubers, the seeds, and so on (Botto 1840, 465). However, argued Botto, the globules derived from pollen had a vivacity of motion not encountered in globules derived from other parts of the plant. Such vivacious motion, he claimed, qualified as the effect of a spontaneity which was peculiar to animal nature. Botto proceeded to investigate the influence of various chemical substances and physical agents on the movement of organic globules suspended in water. He found that a small quantity of ammonia killed almost all movement. Sulfuric, nitric, hydrochloric acids, and opium produced similar deadening effects. The application of strong heat and electricity on the suspending liquid also immobilized the moving globules (Botto 1840, 462).

Contrary to Brown, Botto claimed that the movement of suspended inorganic particles had different features from that of organic globules: "Neither powdered glass, neither quartz, nor the granite of our Alps, nor the pebbles of our rivers, nor rocks of any kind, offered particles endowed with movements analogous to those of organic globules. I could not either certify their presence anymore in the organic substances after carbonization or incineration."<sup>16</sup> The explanation of the movement of the

die Bewegung für rein mechanisch, und zwar durch ungleiche Temperatur des stark erleuchteten Wassers, durch Verdampfung desselben, durch Luftzug und Wärmeströmung u. s. w. Erzeugt" (Muncke 1829, 161).

<sup>&</sup>lt;sup>14</sup> "Au milieu de ces contradictions, et dans un sujet aussi important et complexe, ce qu'il y a de mieux à faire, est de multiplier les expériences, sans franchir trop à la légère les limites de l'observation" (Botto 1840, 459).

<sup>&</sup>lt;sup>15</sup> "On les voit changer à chaque instant de position relative, s'approcher, s'éloigner, tournoyer, comme ci ces mouvements venaient de leur propre fait" (Botto 1840, 459).

<sup>&</sup>lt;sup>16</sup> "Ni le verre pilé, ni le quartz, ni le granit de nos Alpes, ni les cailloux de nos rivières, ni les roches de toute espèce ne m'ont offert de globules doués de mouvements analogues à ceux des globules végétaux. Je n'ai pas pu en constater non plus la présence dans les substances végétales après la carbonisation ou l'incinération" (Botto 1840, 466-67).

inorganic particles by familiar mechanical causes seemed to him to be "neither impossible nor difficult" (Botto 1840, 467). On the other hand, the movement of the organic globules could not be explained by known physical causes. It had, therefore, to be considered a proper quality of the globules themselves, and of their organic and vital nature (Botto 1840, 468). Botto's research shows that vitalist claims, although they were distrusted by most researchers, remained a viable option, at least for the movement of organic particles. Although Botto's observations did not seem to have exerted any influence on subsequent research on Brownian movement, they are important from a historiographical point of view, because, once again, they reveal the difficulties surrounding the application of the varying the circumstances strategy for reaching consensus regarding the causal influence of various factors on the phenomenon.

One of the most widely accepted explanations of Brownian movement during this period was offered by Felix Dujardin (1801-1861). Although he used similar methodological reasoning, Dujardin reached entirely different conclusions from Botto regarding the generality of the phenomenon, the influence of physical agents such as heat and electricity, and its cause. Dujardin's remarks on the phenomenon were offered in his influential treatise on microscopy, in a chapter titled "Du Mouvement Brownien ou Mouvement Moléculaire" (Dujardin 1843, 58-60). The latter followed a chapter devoted to expounding some of the main causes of illusions and errors occurring in microscopy observations. It seems that the existence of disagreements regarding the basic features and causes of the phenomenon invited reflection about possible sources of error. Dujardin cited the invariance of the phenomenon to the influence of various physical and chemical agents-such as light, electricity, magnetism, chemical reagents-to argue that the movement was a purely physical phenomenon belonging to all particles of solid matter that were sufficiently small to be suspended in liquids. In fact, he wanted to warn the uninitiated observer who might perceive in it the manifestation of life and all other kinds of organic activity (Dujardin 1843, 59-60). Studying oil globules suspended in milk, Dujardin found that the vivacity of the movements depended on the size of the particles. The smallest particles, of radius less than 1/600mm, moved the most vigorously, those of radius between 1/400 to 1/300 mm exhibited a movement that was noticeable only if one observed carefully, whereas those of larger size remained motionless. He also found the movement to be more vivid the less the density of the material from which the suspended particles were derived was from that of the water (Dujardin 1843, 59). Dujardin claimed heat to be the only physical agent which affected the phenomenon: it caused the movements to become more rapid. Reflecting on these experimental facts, he was led to conclude that the movements of the suspended particles could be attributed to the various impulses that each particle receives from the radiant heat emitted by the particles adjacent to it.<sup>17</sup>

Dujardin's views on the cause of Brownian movement were shared by Griffith and Henfrey in Britain and were included in their *Micrographic Dictionary* (Griffith and Henfrey 1856). Like Dujardin's treatise, the Dictionary too began with a methodological introduction concerning the proper use of microscopes and the main sources of errors related with their employment. The remarks on Brownian motion were included in the entry *Molecular Motion*—where the term molecule is used to refer to extremely minute particles of any substance. Although the entry gives the impression that it was based on original experimental work, it was in fact a summary of Dujardin's text, with the part referring to the probable causes of motion being simply the English translation of Dujardin's words.<sup>18</sup>

Jules Regnauld (1822-1895), physics professor at the Ecole de Pharmacie in Paris, cited extensive experimental work on the phenomenon, to conclude, in 1858, that Brownian movement was caused by the solar heat absorbed by the suspended particles. When transferred to the surrounding liquid, this heat created very small currents which were responsible for the observed motions.<sup>19</sup>

<sup>&</sup>lt;sup>17</sup> "si l'on chauffe le liquide, le mouvement devient notablement plus vif, et comme tout autre agent physique ou chimique, la lumière, l'électricité, le magnétisme, le contact des réactifs chimiques ou des divers solides est sans influence sur le mouvement Brownien, on est conduit à penser que c'est le résultat des impulsions variées que chaque particule reçoit de la part du calorique rayonnant émis par tous les corps voisins" (Dujardin 1843, 59-60).
<sup>18</sup> "Heat is the only agent which affects it [molecular motion]; this causes the motion to become more rapid. Hence it may be attributed to the various impulses which each particle receives from the radiant heat emitted by those adjacent" (Griffith and Henfrey 1856, 429).

<sup>&</sup>lt;sup>19</sup> "M. J. Regnauld est porté à conclure que les oscillations des corps très-divisés nageant au sein d'un liquide diathermane sont dues à leur échauffement par la portion de la radiation solaire que, absorbée par eux, les rend visibles. Cette faible quantité de chaleur se transmettant par voie de de conductibilité au liquide en contact avec les

The investigators of the phenomenon during these earlier phases of experimental research failed to reach an agreement about the essential characteristics of the phenomenon and the influence of the various suspected causal factors. To clarify and better understand the nature of the disagreements, it would be useful to distinguish between *causal claims* and *causal explanations* made regarding the causal origin of Brownian movement.<sup>20</sup> A causal claim asserted the identification of a "difference-maker", i.e., the causal influence of a suspected factor—such as evaporation, heat, electricity, and so on—on the movement of the suspended particles. By changing or varying the suspected causal factors, the experimental strategy of varying circumstances aimed at identifying such a difference maker and thus making a causal claim. A causal explanation of Brownian movement aimed at providing an—more or less detailed—account of a concrete mechanism linking the causal factor with the effect, i.e, the observed Brownian movements. A causal explanation was more speculative than a causal claim since its details could not be established by using the strategy of varying the circumstances. The latter, however, could be used to identify the difference-maker which could then be used to offer a probable causal explanation of the observed movements.<sup>21</sup>

The early experimental investigations of Brownian movement failed to reach a consensus regarding the identification of a difference-maker. This was to be expected given the difficulties surrounding the application of the varying the circumstances strategy on such a complex phenomenon. Even when some agreement was reached on the influence of some (macroscopical) agent—such as heat—on the movement of the suspended particles, researchers disagreed about the exact mechanism by which this agent, at the microscopical level, produced the observed movements. In the rest of this chapter, we see various permutations of the relationship between causal claims and causal explanations that emerged in the nineteenth century investigations of Brownian movement.

#### 4. Non-Molecular Causal Claims and Explanations of Brownian Movement: 1860-1880

The explanation of Brownian movement by the absorption and radiation of heat turned out to be quite popular. In Britain, a prominent defender of this view was John Benjamin Dancer (1812-1877), microscopist from Manchester. Dancer claimed his conclusions to be based on numerous experiments performed over the course of thirty years with various substances and solutions (Dancer 1868, 162). Dancer claimed that the intensity of the movements depended on the size and shape of the particles as well as on the nature of the solutions. The particles approaching a spherical shape usually exhibited a more marked movement. To further support his claim, Dancer excluded chemical and electrical influences from causing the phenomenon. This he did by demonstrating that the particles did not show any marked alterations in their movements when exposed to electric and chemical influences (Dancer 1868, 164; Jevons 1870, 83).

Dancer claim went against another popular claim regarding the causal origin of the phenomenon in Britain at the time, which presented Brownian movement mainly as an electric phenomenon. The most prominent defender of this claim was William Stanley Jevons (1835-1882), the British philosopher and polymath. Jevons coined the name *pedesis* from the Greek  $\pi\eta\delta\eta\sigma\iota\varsigma$  (meaning leaping or bounding), and the adjective *pedetic* from  $\pi\eta\delta\eta\tau\iota\kappa\delta\varsigma$ , as more appropriates for describing the dancing movement of the suspended particles. The term *molecular movement* used by Brown was inadequate because the moving particles were not molecules in the new chemical sense, whereas the term *Brownian movement* was an inconvenient two-word expression which, in addition, concealed the fact that Brown was not the first to observe the phenomenon (Jevons 1878, 171). Jevons too claimed that his conclusion was the result of extended experimental investigations on the phenomenon (Jevons 1870).

In endeavoring to discover its cause, Jevons conducted a certain number of observations and experiments to test the validity of the various causal claims (i.e., claims in the sense of identifying a difference maker) that had been offered. First to be tested and disproved was the claim upheld by Dancer

particules semblé la cause de petits courants rendus manifestes par les changements de position relative des substances tenues en suspension" (Chatin 1858, 141).

 $<sup>^{20}</sup>$  In making this distinction, we are following Russo and Williamson (2007), who claim that a causal connection can be established only if it can be established (a) that there is a difference-making relationship between the cause and the effect, and (b) that there is a mechanism linking the cause and the effect that is responsible for such a differencemaking relationship.

<sup>&</sup>lt;sup>21</sup> See previous footnote.

and others, that the movement was caused or excited by light or heat falling upon the liquid. Working with particles derived from substances such as kaolin (or china clay as it was commonly called at the time), road dust, red oxide of iron suspended in distilled water, Jevons found that their vibratory movements proceeded exactly in the same manner both in comparative darkness and in intense light from the sun. The movements showed no apparent change even when differently colored glass screens were interposed between the liquid and the sunlight (Jevons 1878, 172). The same conclusion was obtained by means of a comparative experiment. Two suspensions of china clay in water were taken, one was placed in a dark place, while the other was exposed to the direct rays of the sun for three hours. No difference in the rapidity of subsidence of the particles was perceived (Jevons 1878, 172). Regarding the influence of heat in particular, Jevons reached surprising, entirely opposite conclusions to those of previous investigators. More specifically, he was led to believe that the increase of temperature decreased the motion. Jevons did not perceive any difference in the movements of the suspended particles when he warmed the microscope plate. He then tried a comparative experiment. A mixture of charcoal-powder and boiled water was surrounded with ice, while a similar mixture was placed in boiling water and maintained at 100° C. At the end of the hour the heated mixture had deposited nearly all the charcoal, whereas the ice-cold water had as much in suspension after eight hours. A similar experiment with suspensions of china clay gave similar results. Trying to explain the surprising results, Jevons surmised that it was produced by the increase of electrical conductivity of liquids caused by the rise of temperature (Jevons 1878, 173).

Jevons called the comparative experiments described above 'indirect', but this was not because he perceived any difference concerning their epistemic import compared to traditional experimental intervention, where the comparison is between the situation before and the situation after an intervention (or a variation of a circumstance) is made. He called them 'indirect', because the comparative experiments rather than investigating the effect that light and heat had on the particles' vibratory movements, they were investigating the effect that these agents had on the particles' rate of subsidence. In other words, Jevons ascertained the association of pedesis with the suspension of particles in water, and then performed comparative experiments which investigated the influence of various factors on particles' suspension (as opposed to their movements).

Jevons' comparative experiments, however, differed from traditional experimental interventions (or variations) with respect to their epistemic role.<sup>22</sup> Jevons used the comparative experiments to investigate the longer-term effect(s) of the change or variation of the suspected cause (as opposed to its instantaneous or immediate effect(s)). This difference in epistemic role is demonstrated by another (indirect) comparative experiment which convinced Jevons that no external causes to the suspending liquid were involved in the production of pedesis. Trying to test the effect of light and heat on the phenomenon, Jevons took a suspension of china clay in water, and frequently heated it in the fire for two days, allowing it to cool during the intervals. An exactly similar suspension was sunk in sawdust which had been lying for several years undisturbed in a wine-cellar. After remaining for fifty-two hours in complete darkness at a constant temperature of 9° C, the second preparation was found to contain more clay in suspension than the first one which had been moved about and heated many times. Even after seven days the buried preparation "showed a slight cloudiness" (Jevons 1878, 173).

Another time-sensitive question put to the test was whether pedetic motion exhausted itself rapidly or it was retained for long periods of time. Jevons found that ink which was many months— possibly years— old exhibited the motions in perfection. A slow, distinct motion of suspended particles was observed in a drop of lees from a bottle of wine which had been lying undisturbed in a wine cellar for several years. The drop was placed under the thin glass cover of the microscope, with the least exposure to air. The motion was not increased when some of the dregs were shaken up in a bottle with air. The most surprising and conclusive fact of this investigation, however, came from a comparative experiment. In this experiment old mixtures of china clay and water were compared with fresh ones. Two glass tubes containing china clay and distilled water were laid in a drawer for a long period of time. The drawer was usually opened several times in a day, so the tubes would be shaken up every now and then. Frequently the two tubes were shaken by hand. At long intervals the old tubes were opened and drops of the milky liquid were examined. Comparing the motion of the suspended china clay particles in the old mixtures

<sup>&</sup>lt;sup>22</sup> In her contribution to this volume, Schürch also discusses how eighteenth-century researchers investigating the influence of electricity on plant growth perceived the difference between comparative and traditional, interventive experimentation. See also Bernard 1856, 80-2.

with the motion of newly mixed particles found that "*no diminution of motion was apparent*; on the contrary, *the motion seemed to be even more remarkable than in a fresh mixture*" (Jevons 1878, 174; italics in the original). This comparative trial lasted for nine years and led Jevons to declare pedetic motion "the best approach yet discovered to perpetual motion" (Jevons 1878, 174).<sup>23</sup>

To investigate the relation of the movement with the shape of the particles, Jevons compared in a microscope "the fine needle-shaped particles of asbestos dust with the spherical globules of milk, the minute spheres of gamboge, the flat particles of talc, the small cubes of galena, and the wholly irregular fragments of glass." Since all the differently shaped particles exhibited pedesis, Jevons concluded that no particular shape was essential to its production. Contrary to Dancer, however, he found that, *ceteris paribus*, sharp-pointed and irregularly shaped particles oscillated more quickly than spherically shaped particles (Jevons 1878, 173-74).

Jevons considered inconclusive all the experiments which rejected the relevance of electricity for pedetic motion because external electrical currents applied on the liquid had no effect on the movements of the suspended particles. Jevons conclusion that pedesis was due to electricity was based on experiments that put more weight on the variations of the chemical nature of the suspending liquid. He did not learn much by varying the nature of the suspended particles, finding that particles from substances of the most different chemical character exhibited similar pedetic motion (Jevons 1870, 78; 1878, 176). In varying the chemical nature of the liquid, by dissolving various substances therein, however, he discovered that only the purest distilled water gave the movements in their highest perfection. With a few exceptions, all acids, alkalis, or salts tended to diminish the movement, but in a manner that was wholly independent of their peculiar chemical qualities, and dependent only on their electric properties (Jevons 1870, 79; 1878, 179). More specifically, what convinced Jevons that pedesis was due to electric action was the close analogy between his findings when varying the chemical nature of the liquid and the circumstances in which electricity was produced by the hydro-electric machine. Only pure water produced the most amount of electricity in the hydro-electric machine, and almost any salt, acid, or alkali prevented production by rendering the water a conductor (Jevons 1870, 79-80).<sup>24</sup> Pure caustic ammonia, a substance which remarkably did not render water a good conductor and did not prevent the hydro-electric machine from giving electricity was used in a crucial experiment. Jevons dissolved ammonia in water in different amounts and found that it had no effect on the movement of the microscopic, suspended particles (1870, 79-80). Jevons emphasized the fact that his conclusions were based on a great number of experiments done with suspended particles derived from different substances, and involved a great number of substances that were dissolved in the suspending water in various amounts. All the variations in the chemical nature of the suspending liquid, with only few "doubtful exceptions", showed that dissolved substances that turned the water into a conductor of electricity, also inhibited pedetic motion. Jevons distinguished his causal claims regarding the relevance of electricity for the phenomenon-which he regarded as more or less certain since they were based on a large number of observations and experiments<sup>25</sup>—from his more speculative explanations regarding the mechanism of

<sup>&</sup>lt;sup>23</sup> In *Against Method* ([1975] 1993), Paul Feyerabend used the example of "Brownian motion" to support the claim that empirical facts are not simply 'given' but that the description of every single fact depends on *some* theory, and that, in addition, some empirical facts cannot be unearthed except with the help of alternative theories to the theory to be tested. More specifically, Feyerabend claimed that *without the introduction of the kinetic theory*: (a) it is not clear whether the relevance of Brownian motion for the phenomenological second law of thermodynamics could have been discovered, and (b) it is certain that it could not have been demonstrated that Brownian motion actually *refutes* the phenomenological second law (Feyerabend ([1975] 1993, 27). Jevons' longer-term comparative experiments show that the relevance of Brownian movement for the phenomenological second law could be perceived without considering the kinetic theory. In addition, as we show in this chapter, the nineteenth century investigations of Brownian movement, which ended up demonstrating the persistence of the phenomenol despite the variation of the external to the suspending liquid circumstances, make it less certain that an experimental investigation of Brownian movement could not, by itself, pose a challenge to the phenomenological second law.

<sup>&</sup>lt;sup>24</sup> "The analogy of these circumstances to those of pedesis is so remarkable that little doubt can be entertained that the same explanation applies. *It is perfectly pure water which produces electricity and pedesis*. Almost all soluble substances prevent both one and the other; but ammonia is one of a few exceptions – it allows both electric excitation and pedesis. Boracic acid is another exception, and gum a third one" (Jevons 1878, 182; italics in the original). <sup>25</sup> "My recorded observations amount to nearly eight hundred, and the solutions named were tried not only in different strengths, varying according to circumstances, from one part in ten to one part in a million, but they were tried with various suspended powders, such as charcoal, red oxide of iron, amorphous phosphorous, precipitated

electric action on the suspended particles. More specifically, regarding the exact *modus operandi* of the electric action Jevons speculated that it was probably connected with the phenomenon of electric osmose (Jevons 1878, 183).

In a later series of experiments, Jevons used solution of common soap to decide between the causal claim of electric action and the newly proposed claim which asserted that pedesis was caused by surface tension in water. (Jevons 1878, 175; 1879). Soap could serve as a crucial substance for deciding between the two alternative claims because it considerably reduced the surface-tension of water in which it is dissolved, without affecting its electric conductibility. If pedesis was caused by surface-tension, reasoned Jevons, then the motion of the suspended particles would be destroyed or diminished when soap was dissolved in the suspending water. The experiment was tried with particles derived from china clay, red oxide of iron, chalk, barium carbonate, etc., and it gave the opposite result: the pedetic motion of the suspended particles appeared to be increased or facilitated. For Jevons the experiment constituted further proof that pedesis was a phenomenon of electric origin, appearing only in liquids of high electric resistance (Jevons 1879, 435).

Jevons' conclusions regarding the cause of pedesis were challenged, in turn, by William Ord. Ord preferred retaining the term 'Brownian movement', since "everyone knows at once knows what is meant when Brownian movements are spoken of, and, what is of no little importance, the term is extensively used in the continent" (Ord 1879, 656). Although not aware of Jevons' experimental work before its publication, Ord claimed to have independently repeated and confirmed some of his experimental findings such as the hindering action of acids on the movement of the suspended particles (Ord 1879, 658-60). While he admitted that heat, electricity, capillary action, water's surface tension, chemical and other forces may each or all play a part in producing Brownian movements, Ord claimed its main cause to be "vibrations or intestinal disturbances in the colloid suspending fluid, such as attend its decomposition, or its metamorphosis or its resolution into a crystalloid" (Ord 1879, 658).<sup>26</sup>

This conclusion was based on a reasoning not dissimilar to Jevons'. Ord found that the Brownian movements were more active and persistent under conditions which favored the activity of chemical changes in the suspending fluid, and, conversely, that the movements were diminished or altogether stopped by the introduction of conditions which hindered such chemical reactions. Ord explicitly stated that he used-what Mill in the meantime had named as-the method of concomitant variations and the method of difference to support his induction. Regarding the first, he found that "the concomitant variations set forth" showed "that the movement of particles is more or less active according to the presence in the surrounding fluid of conditions favouring or hindering chemical changes in the colloid" (Ord 1879, 660). Ord claimed he used the method of difference in studying mixtures of India-ink with distilled water.<sup>27</sup> When the solid ink was rubbed gently with water, a mixture of suitable thickness was obtained consisting of particles of solid black matter suspended in water which was now dissolving the colloid matter which was binding the ink particles. On the other hand, when a large quantity of ink was rubbed down with water, and the mixture was left in a tall vessel to allow the subsidence of the particles, the colloid matter was gradually washed away, leaving a mixture of particles with nearly pure water. When compared to particles of the same size and number in the first mixture, particles in the second mixture showed an infinitely less active and persistent movement (Ord 1879, 660).

Finally, Ord reinterpreted Jevons's experiments with solutions of soap in a way that supported his own conclusion. Whereas for Jevons the introduction of soap into the suspending fluid increased the movements of the suspended particles because soap retained or did not conduct electricity, for Ord it was a colloid that kept up the movements by revolutionary perturbations (Ord 1979, 660-61).

#### 5. Brownian Movement and Atomic-Molecular Theories of Matter: Early Investigations

According to historian of science Mary Jo Nye, a major reason for the delayed connection of Brownian movement with a molecular conception of matter was that until the second half of the nineteenth century there was no atomic theory of matter capable of offering a suitable mechanism that would causally

carbonate of lime, red oxide of lead, black oxide of manganese, and occasionally with other substances. I don't think, then, that I can be much mistaken in my chief conclusions" (Jevons, 1878: 180).

<sup>&</sup>lt;sup>26</sup> "To sum up...I claim the intestine vibration of colloids as in many cases an agent in the process, and more especially in the fluid and semi-fluid parts of animal and vegetable organisms" (Ord 1879, 662).

<sup>&</sup>lt;sup>27</sup> "I may cite an experiment in which the method of difference gives results in the same direction" (Ord 1879, 660).

connect the atomic-molecular structure of liquids with a phenomenon having the characteristics of Brownian movement. Atomic theories prior to mid-nineteenth century offered a static conception of atoms that interacted with one another primarily through acting-at-a-distance attractive and repulsive forces (Nye 1972, 46; see also Gouy 1895, 5).

Nye is right in pointing out that the explanation of Brownian movement in terms of the molecular motions constituting the liquid state of matter required a molecular theory capable of offering a suitable mechanism explaining how the cause (molecular motions) produced the effect (observed Brownian movements). We should not fail to acknowledge, however, the complexity of the nineteenth century relationship between the ability of making a causal claim regarding Brownian movement and that of providing a causal explanation of it, as pointed out at the end of section 3. So far, we have seen that most nineteenth century investigators of Brownian movement started with the experimental strategy of varying the circumstances aiming to identify a difference maker, i.e., a causal circumstance influencing the phenomenon. In a second step, some of them ventured into speculating about a (more or less) concrete mechanism which, by linking the difference-making circumstance with the observed Brownian movements, was responsible for the experimentally detected difference-making relationship. In the rest of the chapter, we examine some of the permutations of the relationship between causal claims and causal explanations that emerged in the efforts to connect the observed Brownian movements with an atomic-molecular theory of matter during the second half of the nineteenth century.

The first to explicitly connect Brownian movement with an atomic theory of matter was Christian Wiener (1826-1896), professor of descriptive geometry and geodesy at the University of Karlsruhe. In fact, Wiener used the phenomenon of Brownian movement (*Molecularbewegungen*) to provide support for his atomic theory of matter (Wiener 1863). Wiener's atomic theory was a hybrid between the older static conception of atoms and the newer kinetic conceptions which were beginning to emerge at the time. According to Wiener, matter is composed of matter atoms—which attract one another—and aether atoms—which repel one another. The aether atoms are found in the empty spaces between the mutually attracting matter atoms—with aether and matter atoms repelling one another (Wiener 1863, 79). This network of forces exerted between matter and aether atoms meant that matter was in a state of permanent vibration. *Molecularbewegungen*—the trembling motion of microscopic particles suspended in liquids—was then the result of the constant vibrational atomic motions constituting the liquid state of matter (Wiener 1863, 85). Wiener supported his causal explanation of Brownian movement not by providing independent (empirical) evidence for it, but by rejecting other alternative claims about the causal origin of *Molecularbewegungen*.

Lacking positive evidence for his atomic explanation, Wiener used the strategy of varying the circumstances to experimentally disprove, one by one, (all) other alternative causal claims (Wiener 1863, 86). First, Wiener argued, the motion could not be that of infusoria or caused by the vitality of the particles, since he could observe it in finely divided suspended particles derived from inorganic matter. To reject the possibility that the moving particles derived from inorganic substances were actually organic particles trapped in inorganic matter, Wiener annealed quartz particles and found that this had no effect on their movements when suspended in liquids. This same possibility was excluded by the fact that all the suspended particles exhibited the movements as opposed to only few of them (Wiener 1863, 86). Second, the movement was not caused by mechanical, or any other external influences communicated to the suspending liquid. The movements of the suspended particles were more like vibrations, and no one had ever observed such irregular, tremulous movements being caused by external influences. In addition, if the movements were caused by external influences, they ought to change or decrease with time. But Wiener's microscopy observations made over the course of many days revealed an incessant movement which showed no signs of decrease (Wiener 1863, 86). Third, the movement could not be caused by attractive or repulsive forces (electric or other) between the suspended particles, because it was independent of the number of particles present in the liquid and the distances between them. Suspended particles in a dilute emulsion and in relatively large distances from one another exhibited the same trembling motion as that of many particles close together (Wiener 1863, 87). Fourth, the movement could not be caused from temperature differences between the different parts of the liquid. These temperatures differences would offset or decrease with time, whereas the main characteristic of the particles' trembling motion was its invariance through time. In addition, the temperature differences would produce currents from the surface to the interior of the liquid and could not explain the trembling motion of the particles which constantly changed direction even in very small volumes. If the temperature differences were indeed the cause of the trembling motion, the latter had to increase in vivacity when the environment

temperature was changed abruptly. But no changes in the movement were observed despite sudden temperature changes in the surrounding environment (Wiener 1863, 87-89). Fifth, the movement was not caused by evaporation, because evaporation usually takes place near the surface of the liquid, whereas Wiener's microscopy observations revealed that the movement of the suspended particles occurred at all levels of the liquid, and it continued in the same manner even when measures to preclude any evaporation were taken (Wiener 1863, 89-90).

In short, Wiener excluded all the plausible causal claims that could provide the empirical basis for an alternative causal explanation of Brownian movement. He did this by showing that the phenomenon remained invariant, when each one of the suspected causal factors was either varied or entirely excluded from influencing the phenomenon. He concluded that the exclusion of all these suspected difference-makers left no other explanation besides the one attributing Brownian movement to the vibration of the atoms constituting the liquid state of matter: "It remains nothing left but for us to seek the cause [of the phenomenon] in the liquid, and *to ascribe it to the movements constituting the liquid state*".<sup>28</sup>

Another investigator who connected Brownian movement with a mechanical theory of heat was Giovanni Cantoni, professor of experimental physics at the University of Pavia (Cantoni 1867). Cantoni's investigations on the phenomenon, like those of Botto, were ignored by his contemporaries and rediscovered only by the efforts of the historian Icilio Guareschi in the beginning of the twentieth century (Guareschi 1913).<sup>29</sup> Cantoni saw in the phenomenon of Brownian movement (*moto Browniano*) the confirmation of a mechanical theory of heat.

For Cantoni, the heat of a body consists in the vibratory movements of its constituent molecules. Every chemical substance, at a given temperature, has a characteristic vibratory motion of its constituent molecules. This was macroscopically indicated by the fact that different amounts of heat are required to increase by the same degree of temperature the same weight of different substances—i.e., by the existence of the different substances' specific heats.

According to Cantoni's proposed explanation, Brownian movement was caused by the different molecular velocities that must exist at the same temperature between the molecules constituting the solid suspended particles, on the one hand, and the molecules of the suspending liquid hitting the suspended particles from every direction, on the other.<sup>30</sup> Cantoni argued that this explanation could be experimentally tested and positively confirmed: ceteris paribus, Brownian movements ought to be more vivid the greater was the difference between the velocities of the molecules constituting the solid particles from the velocities of the molecules constituting the suspending liquid. At the macroscopical level, the difference between the molecular velocities of different substances was simply the difference between their specific heats (Cantoni 1867, 163). If the difference between molecular velocities was the real cause of Brownian movements, then varying the difference between the specific heat of the suspended particles and the specific heat of the suspending liquid ought to bring a corresponding variation in the intensity of Brownian movements. Cantoni claimed that his numerous experiments performed with various suspended particles and suspending liquids showed that this was indeed the case. For example, particles derived from the same substance moved far more intensely in water than in alcohol. Since alcohol has a lower specific heat than water, there was a smaller difference between the specific heat of the suspending liquid from that of the suspended particles. Following similar reasoning, one could explain why the Brownian movement of identical particles was even less marked in gasoline and ether than in water (Cantoni 1867, 163-67). All this evidence, according to Cantoni, concurred to the conclusion that the cause of the phenomenon resided in the different velocities that the molecules of different substances have at the same temperature. From this evidence, Cantoni jumped to the conclusion that the existence of Brownian movement provided one of the most beautiful and direct experimental demonstrations of the fundamental principles of the mechanical theory of heat, manifesting the assiduous vibratory state that must exist both in liquids and solids even when their temperature does not change.<sup>31</sup>

<sup>&</sup>lt;sup>28</sup> "[E]s bleibt uns daher Nichts übrig, als die Ursache in der Flüssigkeit an und für sich zu suchen, und sie *inneren dem Flüssigkeitzsustande eigenthümlichen Bewegungen zuzuschreiben*" (Wiener 1863, 90; italics in the original).

<sup>&</sup>lt;sup>29</sup>According to Guareschi (1913: 50), Cantoni was the first to clearly discover the true cause of the phenomenon. <sup>30</sup> "Ebenne, io penso che il moto di danza delle particelle solide estremamente minute entro un liquido, possa attribuirsi alle differenti velocità che esser devono ad una medesima temperatura, sia in codeste particelle solide, sia nelle molecole del liquido che le urtano d'ogni banda" (Cantoni 1867, 163).

<sup>&</sup>lt;sup>31</sup> "Ora tutti gli esposti particolari concorrano alla deduzione, che la condizione fisica del moto browniano stia nella diversa velocità che hanno le molecule dei corpi differenti sotto una stessa temperatura. E di tal modo il moto browniano, così dichiarato, ci fornisce una delle più belle e dirette dimostrazioni sperimentali dei fondamentali

Wiener's atomic explanation of Brownian movement was based on the rejection of all other alternative causal claims. The rejection of all the possible macroscopical difference-makers, left no other explanation than the one attributing the movement of suspended particles to the vibratory movements of aether and matter atoms which, according to Wiener's atomic theory, constituted the liquid state of matter. Embedded as it was in an idiosyncratic theory of matter which had no independent empirical evidence in its favor, Wiener's explanation was deemed inadequate. On the other hand, Cantoni's explanation in terms of the different molecular velocities that, according to his molecular theory of heat, must exist at the same temperature between the molecules of the suspended particles and the molecules of the suspending liquid, manifested itself in a macroscopical difference-making relationship which could be experimentally manipulated to provide empirical support. Cantoni's work, however, did not receive any attention and thus did not have any influence on subsequent research on the phenomenon (Guareschi 1913). To my knowledge, even the difference-making relationship detected by Cantoni was not replicated by someone else. One possible reason for the neglect of Cantoni's explanation, must have been his peculiar mechanical theory of heat which contradicted some of the basic tenets of the newly developed, and more successful, kinetic-molecular theory (see next section). The main obstacle facing all (kinetic-) molecular explanation of Brownian movement during this period, however, was the emergence of arguments which challenged the adequacy of the hypothesized molecular motions for causing a phenomenon with the observable characteristics of Brownian movement (Nye 1972, 23; Nägeli 1879; Ramsay 1882).

#### 6. Brownian Movement and the Kinetic-Molecular Theory of Matter

In this section we examine the reasoning of the researchers who were the first to explicitly connect the phenomenon of Brownian movement with the thermo-dynamic motion of molecules as proposed in the then newly developed kinetic theory of gases. These were a group of Jesuit scholars associated with the journal *Revue des questions scientifiques*, published by the Scientific Society of Brussels (Nye 1976). Rather than starting with varying the circumstances to exclude alternative causal claims and/or identify difference-makers, the proponents of the kinetic-molecular explanation tried to show that the tenets of the kinetic-molecular conception of matter which were developed independently to explain a different range of observable phenomena—namely the macroscopic behavior of gases and liquids—could provide a causal explanation for the altogether different phenomenon of Brownian movement. The ability of the kinetic-molecular theory to account for a range of unrelated phenomena and experimental evidence, was used to "control" its validity as well as the validity of the offered explanations.<sup>32</sup>

The first explicit connection of Brownian movement with the kinetic theory of gases was made by Father Joseph Delsaulx, a Brussels-born Jesuit, in a paper whose aim was to show: "that all the Brownian motions of small masses of gas and of vapour in suspension in liquids, as well as the motions with which viscous granulations and solid particles are animated in the same circumstances, proceed necessarily from the molecular heat motions, universally admitted, in gases and liquids by the best authorized promoters of the mechanical theory of heat" (Delsaulx 1877, 2).

Delsaulx provided a detailed account of how the invisible molecular motions postulated in the kinetic theory of heat to explain the macroscopic behavior of gases, would cause the dancing movement of microscopic particles suspended in liquids. More specifically, it followed from the principles of the mechanical theory of heat that a favorable concourse of the movements of oscillation, rotation, and translation of the molecules of the suspending liquid would, by necessity, produce on isolated points on the surface of a suspended particle a pressure of an exceptional intensity. These pressures were averaged out in particles of larger dimensions, but not in the microscopical dimensions of Brownian particles. They were thus the real cause of the particles' continuous oscillatory motions (Delsaulx 1877, 3-6). "All these [Brownian] movements," Delsaulx concluded, "result from the interior dynamic state that the mechanical theory of heat attributes to liquids, and are a remarkable confirmation of it" (Delsaulx 1877, 5).

The kinetic-molecular explanation of Brownian movement could make sense of the various observed features of the phenomenon. Namely that Brownian movement was more active in heated

principii della teoria mecanica del calore, manifestando quell'assiduo stato vibratorio che esser deve e nei liquidi e nei solidi ancor quando non si muta in essi la temperatura" (Cantoni 1867, 167).

<sup>&</sup>lt;sup>32</sup> This way of reasoning is like that we encounter in William Whewell's (1847; 1858) notion of the *consilience of inductions*. See also Coko (forthcoming).

liquids than in those of a low temperature; that supposing equal diameters, the oscillatory displacement is more rapid and more extended in fatty granulations than in metallic granulations whose density is very great; and the fact that the duration of the phenomenon may be said to be without limit, since it has been observed in gas-bubbles imprisoned in microscopic (liquid-filled) cavities of quartz for supposedly millions of years (Delsaulx 1877, 2).

Another Belgian Jesuit, Julien Thirion, in a lengthy paper published in 1880, similarly argued that Brownian movement could be easily explained by recourse to the mechanical theory of heat. According to the latter, explained Thirion, all bodies are composed of molecules in a perpetual state of motion. Although these molecular motions cannot be directly observed, there was a variety of phenomena and surprising experimental facts that could be easily explained by appeal to their existence (Thirion 1880, 6). For example, the new and surprising experimental facts established in William Crookes' experiments on cathode rays could be readily explained by the tenets of the of the kinetic-molecular conception of gases as proposed in the mechanical theory of heat. What made this explanation even more remarkable, Thirion claimed, was the fact that the kinetic-molecular conception of gases was originally developed to explain a totally different range of phenomena—namely the macroscopic behavior of gases. The simplicity with which the kinetic-molecular conception accounted for these unexpected facts, the fruitfulness of the insights it suggested, and the variety of evidence it predicted and explained, gave the conviction that one was not mistaken in taking it as a guide.<sup>33</sup>

Thirion used Brownian movement as another example of a surprising phenomenon that could be easily explained by the tenets of the mechanical theory of heat. Thirion explained that the latter predicted that sufficiently small particles suspended in water would be in a state of permanent oscillation. According to the mechanical theory of heat, the surface of a solid body suspended in a liquid is continually and unequally bombarded by the movement of the invisible molecules constituting the liquid state of matter. In large particles with sufficiently large surfaces the inequalities of molecular collisions would compensate one another. In these particles, the molecular collisions, despite their high irregularity, would produce no visible effects. In very small particles with a surface sufficiently small to not allow for the compensation of the irregularities, however, the total pressure exerted at any moment from the molecular collisions would no longer be zero but vary continuously in intensity and direction. The center of gravity of the particle would be continuously displaced, the result being the continuous oscillation of the particle. The inequalities in pressure and the oscillations would be more and more apparent the smaller the suspended particles (Thirion 1880, 43-45). For Thirion, the phenomenon of Brownian movement was a remarkable empirical verification of this prediction of the kinetic-molecular conception of liquids. What made the prediction even more remarkable was the fact that the molecular conception of liquids was not developed to accommodate this kind of phenomenon. It was a happy coincidence that such a phenomenon could be detected experimentally. <sup>34</sup>

## 7. Brownian Movement and the Kinetic-Molecular Theory of Matter: Controlling the Evidence and the Kinetic-molecular Hypothesis

The French physicist Louis Georges Gouy (1854-1926) is widely credited for being the first to firmly connect Brownian movement with the molecular motions postulated by the kinetic-molecuar theory of matter.<sup>35</sup> In this section we show that Gouy's success can be attributed to the fruitful combination of the

<sup>&</sup>lt;sup>33</sup> "Si cette science maîtresse avait encore besoin de preuves, il nous semble qu'elle les trouverait ici solides et nombreuses. La simplicité avec laquelle elle rend compte de ce grand nombre de faits inattendus, la fécondité des aperçus qu'elle suggère, la variété des détails qu'elle prévoit et qu'elle explique, donnent à l'esprit la conviction qu'il ne s'est point fourvoyé en la prenant pour guide" (Thirion 1880, 39).

<sup>&</sup>lt;sup>34</sup> "ce ne sont pas des phénomènes qui se présentent à nous et qu'il faut expliquer, *ce sont des conséquences d'une théorie édifiée pour expliquer d'autres phénomènes.* Si l'expérience venait à montrer que ces conséquences ne se vérifient pas, il en faudrait conclure que la théorie est au moins inexacte, peut-être tout à fait erronée. *Heureusement* l'expérience fait tout le contraire" (Thirion 1880, 41-42, italics added). Also, "Tous ces faits, observés par R. Brown, peuvent vraiment être considérés comme une vérification anticipée d'un théorème trouvé un demi-siècle plus tard" (Thirion 1880, 50). <sup>35</sup> "On the contrary, it was established by the work of M. Gouy (1888), not only that the hypothesis of molecular agitation gave an admissible explanation of the Brownian movement, but that no other cause of the movement

could be imagined, which especially increased the significance of the hypothesis. This work immediately evoked a

experimental strategy of varying the circumstances with the theoretical and hypothetical reasoning on the causal origin of the phenomenon. More specifically, Gouy (*a*) used the invariance of Brownian movements to the variation of various suspected factors to reject the claims placing the cause of the phenomenon to influences external to the suspending liquid, and (*b*) showed how the hypotheses regarding the internal constitution of liquids that were independently developed in the context of the kinetic theory of matter, and which were already employed successfully to explain a variety of phenomena, were sufficient to explain the experimental facts of Brownian movement.

Gouy conducted extensive experiments on the phenomenon during the late 1880s and was able to conclusively establish its essential features. He presented the results of this early experimental work in a short note published in the *Journal de Physique* (Gouy 1888). Gouy claimed that Brownian movement was a characteristic of all microscopic solid particles suspended in liquids. Initially he worked with suspensions of gamboge and China ink in water. The water drop containing the particles was covered with a cover slip, and the preparation was enclosed with paraffin to avoid evaporation and external influences. Using an immersion lens Gouy observed a striking trembling motion of the suspended particles. Every particle seemed to move independently of its neighboring particles. Every particle experienced a series of displacements difficult to describe because they were essentially irregular. In particles with elongated form or some mark in their surface, Gouy was able to detect an irregular rotational movement. The movements were more vivid the smaller the size of the particles, they increased with temperature, and were more active in less viscous liquids (Gouy 1888, 561-62).

The careful observation of the phenomenon left no doubt, according to Gouy, that the movements were not the result of vital forces, external vibrations, temperature differences, or other accidental currents in the liquid, but a normal phenomenon, occurring at a constant temperature, and due to the internal constitution of liquids. The independence of the movements from the nature of the particles, their irregular nature, their persistence in time even when all precautions to exclude any external influences were taken, showed their cause to be the internal agitation of the liquid. Brownian movement, therefore provided a 'direct and visible' proof of the molecular-kinetic hypotheses regarding the nature of heat: "Brownian movement, therefore, shows us, of course not the movement of molecules, but something very close to it, and it provides us a direct and visible proof of the correctness of current hypotheses on the nature of heat. If one adopts these views, the phenomenon, whose study is long from over, surely takes a higher order of importance for molecular physics."<sup>36</sup>

Gouy's (1889; 1895) next two papers on the topic, present his experimental strategy and theoretical reasoning in more detail. Gouy experimentally identified the essential characteristics of the phenomenon and inquired into its causal origins. Brownian movement, he remarked, was essentially irregular and seemed to be governed only by chance. It consisted in a series of little impulses which were oriented indistinguishably in all directions, and which were not subject to any law. The movement was some sort of oscillation in place, which nevertheless, in the long run, it could produce noticeable displacements in the position of a suspended particle. The rapidness and amplitude of the movement depended above all on the size of the particles, becoming greater the more the particles were smaller. The movement was not influenced by the form, the state, or the chemical and physical nature of the suspended particles. The movement was more intense in suspending liquids with higher degrees of fluidity. Although the movement of the suspended particles was irregular (with each particle moving independently of its neighboring particles), the phenomenon, taken in its entirety, had an obvious regularity, in the sense that it was always found exhibiting the same essential characteristics (Gouy 1895, 2-3). Gouy claimed to have observed the Brownian movements under the most varied conditions using liquids and particles with different chemical and physical properties without, however, observing any difference in its essential features.<sup>37</sup> Regarding the question of the causal origin of Brownian movement,

considerable response, and it is only from this time that the Brownian movement took a place among the important problems of general physics" (Perrin 1910: 4-5). See also Poincaré 1905, 199.

<sup>&</sup>lt;sup>36</sup> "Le mouvement brownien nous montre donc, non pas assurément les mouvements des molécules, mais quelque chose qui y tient de fort près, et nous fournit une preuve directe et visible de l'exactitude des hypothèses actuelles sur la nature de la chaleur. Si l'on adopte ces vues, le phénomène, dont l'étude est loin d'être terminée, prend assurément une importance de premier ordre pour la physique moléculaire" (Gouy 1888, 563).

<sup>&</sup>lt;sup>37</sup> "Les observations ont été faites avec des particules minérales ou organiques, solides ou liquides, en suspension dans des liquides variés, eau, solutions aqueuses, acides, alcools, éthers, carbures d'hydrogène, essences, etc...D'autres observations ont été faites sur les bulles gazeuses que renfermement les inclusions liquides fréquentes dans certains quartz, et qui sont animées d'un mouvement tout à fait comparable à celui des particules solides ou

Gouy was explicit that it "can only be answered by a detailed study of the phenomenon, under the most varied circumstances possible, by striving to reduce or increase at the outmost limits the external causes of agitation and examining the resulting effects."<sup>38</sup> That is to say, the question could only be answered using the experimental strategy of varying the circumstances.

First, Gouy claimed, that it was easy to show that Brownian movement was not of a vital nature since the phenomenon had been observed in liquids where no living entity could exist: the most toxic substances, acids and the strongest alkalis never stopped the movements; high temperatures which destroyed all life, increased the movements instead of stopping them (Gouy 1895, 2). Second, the generality of the phenomenon and the fact that it seemed to last indefinitely—having been observed in air bubbles suspended in liquids entrapped in cavities of quartz crystals for thousands of years—was sufficient to show that the phenomenon was not due to any external and accidental causes, which had to act with a varying intensity depending on the circumstances (Gouy 1889, 103). To establish this last point decisively, however, Gouy conducted several rigorous experiments. More specifically, to test the various claims about the causal origin of Brownian movement, Gouy examined the change in the essential characteristics of the phenomenon, while the different suspected causes were either excluded or considerably varied. The detailed description he gave of his experiments shows the efforts put to control the influence of external to the suspending liquid disturbances.

The first claim to be tested was whether the Brownian movements were caused by external vibrations communicated to the suspending liquid or undetected tremors coming from the ground. To avoid any external disturbances, the microscopy apparatus was installed in a basement away from any source of agitation. To control for the existence of tremors coming from the ground, or any sort of external vibrations, a basin of mercury was placed next to the apparatus. The surface of mercury acted as a perfect mirror of extreme sensibility for detecting the slightest disturbances. While the surface of mercury remained at complete rest, the Brownian movement continued exhibiting its usual characteristics and intensity; the movement did not increase significantly when external disturbances were noticeable. Based on similar, often repeated, experiments Gouy concluded that external vibrations or ground tremors were not causes of the phenomenon (Gouy 1889, 103-04; 1895, 4).

The second claim to be tested was whether the Brownian movements were caused by currents in the liquid due to temperature differences. Gouy was able to reduce currents caused by temperature differences by immersing the preparation in a water trough, which ensured the attainment of a uniform temperature. To conduct his observations, he used an immersed lens. No variations in the Brownian movement of the suspended particles were observed throughout the entire procedure. In addition, currents in the liquid produced coordinated movements of adjacent Brownian particles that looked nothing like the individual vibrations which constituted Brownian movement (Gouy 1889, 104; 1895, 4).

A third claim to be tested was whether the light required for the microscopy observations, and which passed through the liquid, affected the suspended particles, for example by heating them in an unequal manner. The individual vibrations of the particles would then be the result of such temperature differences. To test this claim Gouy widely varied the nature and the intensity of light used to illuminate the preparation, without observing the slightest difference in the particles' movements. Light, he concluded, played no perceptible role on Brownian movement (Gouy 1889, 104; 1895, 4-5).

Fourth, Gouy contended that other hypothetical causes such as the influence of terrestrial magnetism and electric currents had no influence on Brownian movements, since no variation was observed when the preparation was placed in an electromagnetic field or when electric currents were applied. The only agent that influenced the movement was heat. At temperatures 60° to 70° C, the movement was a little more noticeable than at normal temperature (Gouy 1889, 4; 1895, 5).

Gouy explicitly used the term 'control' to point out the fact that his observations and experimental results could be easily verified independently and were, therefore, independent of any theoretical idea and interpretation.

liquides...Le point le plus important est la régularité du phénomène des milliers de particules ont été examinées, et, *dans aucun cas*, on n'a vu une particule en suspension qui n'offrît pas le mouvement habituel, avec son intensité ordinaire, eu égard à la grosseur de la particule" (Gouy 1889, 103). See also Gouy 1895, 2-3.

<sup>&</sup>lt;sup>38</sup> "A la question ainsi posée, on ne peut répondre que par l'étude détaillée du phénomène, dans des conditions aussi variées que possible, en s'efforçant de réduire ou d'augmenter dans les limites le plus étendues les causes extérieures d'agitation, et examinant les effets produits" (Gouy 1895, 4).

These observations which are easy to control, seem to establish as experimental facts and apart from any theoretical idea: 1<sup>st</sup> that Brownian movement occurs with any kind of particles, with an intensity that is the lesser the more the liquid is viscous and the more the particles are larger, 2<sup>nd</sup> that this phenomenon is perfectly regular, it occurs at a constant temperature and in absence of any external cause of movement.<sup>39</sup>

Leaving the solid ground of observation and experiment, Gouy entered the second part of his argument: that of hypothetical and theoretical reasoning on the causal origin of Brownian movement. Theories and hypotheses, contended Gouy, have been abused and slandered a lot, yet their importance for scientific inquiry is indisputable. They could often shed an unexpected light on a whole set of questions. In addition, the history of the physical sciences showed that theoretical speculations had been the source of the finest discoveries and of the greatest progress. The use of hypotheses was thus legitimate as long as they were used cautiously and were controlled by empirical evidence: "Let's give them their due, the consideration deserved by eminent services, and that limited confidence that never sleeps and does not neglect any means of control."<sup>40</sup>

Gouy argued that the cause of Brownian movement which lasted indefinitely without an apparent cause should not be searched for in the nature of the particles or in any external factors, but in the constitution of the suspending liquid itself. In fact, the hypotheses made in the context of the modern kinetic theory of matter were directly related to the existence of the phenomenon. More specifically, "the kinetic theory could make us predict this phenomenon, and it *explains* it to us in its essential features" (italics added).<sup>41</sup>

After showing how the kinetic-molecular hypotheses could explain the experimentally determined features of Brownian movement, Gouy conceded that the kinetic-molecular explanation faced a problem of underdetermination, namely it assumed that there were not any other unknown causes of which the Brownian movement could be the effect. Gouy maintained, however, that no supposition of such causes was necessary if the kinetic-molecular hypotheses were sufficient to explain the phenomenon. In addition, these hypotheses were not entirely beyond any means of control. They had already led to considerable insights about a variety of physical and chemical phenomena.<sup>42</sup> Among the successes of the kinetic theory Gouy listed the molecular explanations of heat and radiation. Further, the agreement on the numerical values for the molecular dimensions obtained by diverse theoretical methods gave to the claims of the kinetic theory a great aura of plausibility.<sup>43</sup>

To sum up, Gouy used the experimental strategy of varying the circumstances to (a) identify the essential characteristics of Brownian movement, (b) identify macroscopical difference-makers which influenced its intensity—namely heat and the size of the Brownian particles—and (c) exclude other suspected factors from playing a causal role in the production of the phenomenon. The experimental strategy left the kinetic-molecular motions as the only plausible explanation. Although he admitted the existence of a problem of underdetermination, Gouy could appeal to (a) the ability (or necessity, as Gouy saw it) of the kinetic-molecular motions to produce a phenomenon with the observable characteristics of Brownian movement, and (b) the plausibility of the kinetic-molecular conception of matter given its ability to explain a variety of other phenomena. These arguments made unnecessary the appeal to other

<sup>&</sup>lt;sup>39</sup> "Ces observations qu'il est facile de contrôler, paraissent établir comme faits d'experiénces et en dehors de toute idée théorique : 1° que le mouvement brownien se produit avec des particules quelconques, avec une intensité d'autant moindre que le liquide est plus visqueux et les particules plus grosses ; 2° que ce phénomène est parfaitement régulier, se produit à température constante et en absence de toute cause du mouvement extérieur" (Gouy 1889, 104-5).

<sup>&</sup>lt;sup>40</sup> "Accordons leur ce qui leur est dû, la considération que méritent des services éminents, et cette confiance limitée qui ne s'endort jamais et ne néglige aucun moyen de contrôle" (Gouy 1895, 5).

<sup>&</sup>lt;sup>41</sup> "La théorie cinétique pouvait nous faire prévoir ce phénomène, et elle nous l'explique dans ses traits essentiels" Gouy 1895, 7).

<sup>&</sup>lt;sup>42</sup> "La théorie cinétique de la matière a conduit à des aperçus fort intéressants sur un certain nombre de phénomènes physiques et chimiques, et la part qu'elle a prise dans l'œuvre scientifique de notre époque est déjà considérable (Gouy 1895, 6).

<sup>&</sup>lt;sup>43</sup> "C'est aussi la conclusion à laquelle sont arrivés par d'autres voies les physiciens qui ont essayé de se faire une idée des dimensions moléculaires. Par des méthodes diverses, assez concordantes pour qu'on leur accorde crédit, ils sont arrivés à évaluer l'intervalle des molécules dans les liquides à la millième partie environ des dimensions des plus petits corps visibles au microscope. Il faudrait donc environ un milliard de molécules pour former le poids d'une de plus petites particules sur lesquelles nous observons le mouvement brownien" (Gouy 1895, 7).

(unknown) causal factors and thus eased the underdetermination problem. This summary of Gouy's reasoning helps us to make sense of his contention that "Brownian movement provides us with what the kinetic theory of matter was lacking: a direct experimental proof. No doubt, we cannot observe, and we will never be able to observe the molecular movements; but at least we can observe something which results directly from them and necessarily indicates an internal agitation of bodies."<sup>44</sup>

This synthesis of experimental and theoretical modes of reasoning was perfected in Jean Perrin's (1870-1942) experimental work, which established molecular motions to be the *proper and unique* cause of Brownian movement. Perrin was able to determine by means of multiple, independent experiments that the internal motions of the liquid causing the experimentally established characteristics of Brownian movement, were identical with the molecular motions postulated in the kinetic theory of matter. The multiple determination of molecular magnitudes proved to be the ultimate criterion for 'controlling' the veracity of the kinetic-molecular explanation of Brownian movement (Coko 2020a).

#### 8. Summary and Conclusions

In this chapter we have argued that there was an important and sophisticated experimental work done in investigating the main characteristics and causal origin of Brownian movement throughout the nineteenth century. Investigators followed as rigorously as possible the methodological standards of their time to make causal claims and formulate causal explanations about the phenomenon. Two distinct methodological strategies were employed.

The first was the experimental strategy of varying the circumstances. Various suspected causal factors were excluded or varied and the resulting effect on Brownian movements was studied. The main goal of this strategy was the identification of difference-making factors, i.e., of factors having a causal influence on the phenomenon. All factors that could varied or excluded without influencing the movement of the suspended particles were excluded from playing a causal role in its production. On the other hand, all factors whose exclusion or variation influenced the phenomenon were considered to have a role in its production. The identification of a difference-making factor was sometimes followed by theoretical speculation about the concrete mechanism linking the difference-making factor with the observed Brownian movements.

This strategy was already implemented in the first identification and investigations of the phenomenon, at first implicitly, and later, when the initial observations were challenged or led to conflicting results, more explicitly. The strategy of varying the circumstances strategy involved three notions of control: (a) control over the factor to be varied or excluded, (b) control over the rest of the factors which ought to remain constant, and (c) control in the sense of comparing the situation with the varied factor with the experimental situation without it. We can distinguish two types of experimentation employing this strategy. First, there was 'classic' (or indirect) experimental intervention, where the comparison was between the situation before and the situation after the intervention (or variation of the investigated factor). Second, there was comparative experimentation, where the comparison was between these two types of experimentation were made with respect to their underlying rationale and epistemic import, the second kind was used to investigate processes and effects of longer duration—as opposed to instantaneous and immediate effects—or it was used in cases where direct intervention was not possible.

The use of the varying the circumstances strategy did not lead to a consensus regarding the essential characteristics and the causal origin of Brownian movement. The existence of disagreements revealed the importance of another notion of 'control': that of the verification of experimental findings by other researchers, preferably independently from one another. Since most claims regarding the causal origin of Brownian movement could not be verified independently, the varying the circumstances strategy succeeded more in excluding various suspected factors from being causes of the phenomenon rather than in establishing a positive causal claim. Brownian movement proved to be, what today we would call a robust phenomenon, which remained invariant to the variation of most of the experimental factors that

<sup>&</sup>lt;sup>44</sup> "le mouvement brownien nous fournit ce qui maquait à la théorie cinétique de la matière: une preuve expérimentale directe. Sans doute, nous ne voyons pas et nous ne verrons jamais les mouvements des molécules; mais nous voyons du moins quelque chose qui en résulte directement et suppose d'une manière nécessaire une agitation interne des corps" (Gouy 1895, 7).

the experimenters could directly vary and control. Today, with hindsight, we know why. Even when the causal influence of some factor (i.e., difference-maker)—such as heat or size of the Brownian particles on the observed movements received independent confirmation, investigators disagreed on the causal explanation offered, i.e., in their description of the concrete mechanism responsible for the difference-making relationship.

The second strategy used was the hypothetico-deductive strategy or *method of hypothesis*, recognized during the nineteenth century as the proper strategy for validating explanatory hypotheses regarding unobservables. Rather than starting or relying exclusively on the ability of experimental work to identify difference-making factors and/or exclude causal claims, the proponents of the kinetic-molecular explanation tried to show that the tenets of the newly developed kinetic-molecular conception of matter could provide a natural explanation for the essential characteristics of Brownian movement. What was remarkable about this explanation, claimed its proponents, was the fact that the elements of the theory invoked to explain Brownian movement were developed independently, to explain an entirely different range of observable phenomena—namely the macroscopic behavior of gases and liquids. Seen in this vein, the existence of Brownian movement provided unexpected empirical evidence for the kinetic-molecular conception of matter. The ability of the kinetic-molecular theory to account for a range of unrelated phenomena and experimental evidence, was used to "control" its validity as well as the validity of the offered explanation.

None of these two methodological strategies could, on its own, establish molecular motion as the cause of Brownian movement. It was only the fruitful combination of these two—historically antithetical—strategies and their accompanying notions and practices of control that led, at the end of the nineteenth century, to the recognition of molecular motion as the most probable cause of the phenomenon. From then on, the main goal of experimental investigation on Brownian motion became that of evaluating and probing the validity of the kinetic-molecular explanation. This shift in goals brought shifts in the experimental strategies used to establish the validity of claims about unobservable entities and processes such as molecules and molecular motion. Because of these changes, the understanding of what is meant to do "rigorous" experimental research changed as well.

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