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Polarisation en France

Abstract: The special relationship between France and the polarization of light from the beginning of the 19th Century until the present day is reviewed in the lives and works of Étienne Louis Malus, François Arago, Jean-Baptiste Biot, Augustin Fresnel, Louis Pasteur, Frédéric Wallerant, Aimé Cotton, Francis Perrin, and Alain Aspect. To avoid a redundant presentation of information that can be found with an internet search engine, the author emphasizes how the aforementioned individuals have influenced the studies of stereochemistry, molecular chirality, and the polarization of light in his research group.

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1. Introduction

Professor Jeanne Crassous, an organizer of the 16th International Conference on Chiroptical Spectroscopy (Rennes, 2017), asked the author to review the pioneering French contributions to the science of polarized light to honor our gathering's host country. Indeed, France has had a special relationship with polarized light. This paper is an overview of the author's tour of France while riding a beam of polarized light. Presented here is not a complete history of more than two centuries of research, but rather a selection of highlights from the work of individuals whose efforts have influenced the author and his research group over more than two decades.

At the end of his life, Aimé Cotton began to work on a book, *La Lumière Polarisée*. He was not able to finish, but the introduction has been published in a scientific biography written by his wife Eugénie.[1] The most comprehensive history of polarized light was given by a French author, Christian Brosseau,[2] free of national bias. Since the author of the present contribution is not French, and free of French ancestry, to the best of his knowledge, he can emphasize the Gallic contributions at the expense of others without fear of charges of prejudice.

2. Étienne-Louis Malus (1775-1812)[3]

Étienne Louis Malus' is one of 72 *savants* honored on the facade of the Eiffel Tower. "Malus" decorates the Northeast side of the tower, below the first balcony, a placed earned for the discovery of the polarization of light by reflection. We know precisely how the discovery of the polarization of light by reflection was made because it was told by Malus' friend, François Arago, himself honored on Southeast facade (Figure 1).

According to Arago, "[O]ne day in his house in the Rue d'Enfer, Malus happened to examine through a doubly refracting crystal [Iceland spar], the rays of the sun reflected from the windows of the Luxembourg Palace. Instead of two bright images which he expected to see, he perceived only one, -- the ordinary, or the extraordinary, according to the position which the crystal occupied before his eye. This singular phenomenon struck him much."[4] Light had never before behaved in this way that had not already passed through one Iceland spar crystal. Malus had separated the "sidedness" of light from the action of crystals. He had discovered the polarization of light by reflection.

That evening, Malus, working with candlelight in the place solar rays, had deduced the Law of Malus, that the intensity of linearly polarized light transmitted by a linear analyzer varies as the squared cosine of the angle θ between the polarization direction and the preferred direction of the analyzer: $I/I_o = \cos^2(\theta)$.[5] This is the first quantitative expression involving

FIGURE 1. Some of the scientists immortalized on the Southeast side of the Eiffel tower. For Arago, see section 3. Photo by Karl-Heinz Ernst reproduced with permission.

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polarized light, all the more remarkable because the only light detector in 1808 was the human eye. Brightness, perceived by Malus, was compared with standard candles at fixed distances from a white screen.

In our lab, accounting for light polarization is accomplished with the Stokes-Mueller calculus.[6] Stokes vectors describe polarization states of light, and these are transformed from one (**S**) to another (**S'**) by linear operators called Mueller matrices (**M**), Eq. 1. A Stokes vector is built from parameters describing

$$\begin{bmatrix} S_1' \\ S_2' \\ S_3' \\ S_4' \end{bmatrix} = \begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{34} \\ M_{41} & M_{42} & M_{43} & M_{44} \end{bmatrix} \begin{bmatrix} S_1 \\ S_2 \\ S_3 \\ S_4 \end{bmatrix}. \qquad \text{Eq. 1}$$

sums and differences of intensities of light polarized along orthogonal directions, *x* and *y*, directions bisecting those, *x′* and *y′*, and right and left (*R* and *L*) circularly polarized light (CPL). **S** = $[I_x+I_y, I_x-I_y, I_{x'}-I_{y'}, I_R-I_L]^T$ = $[S_1, S_2, S_3, S_4]^T$. Thus,
And, the polarization of unpolarized light, **S**=$[1,0,0,0]^T$, by reflection is reckoned in terms of sums and differences of the angles of incidence and refraction, θ_+ and θ_-, respectively, as shown in Eq. 2, the form of the Mueller matrix of a linear polarizer:

Malus died just a few years after his great discovery at age 37. His health was compromised after contracting the bubonic plague while establishing a hospital for infectious disease in Jaffa, one of his responsibilities as an engineer in Bonaparte's army of Egypt.

$$\begin{bmatrix} S_1' \\ S_2' \\ S_3' \\ S_4' \end{bmatrix} = \frac{1}{2}\left(\frac{tan\theta_-}{sin\theta_+}\right)^2 \begin{bmatrix} cos^2\theta_- + cos^2\theta_+ & cos^2\theta_- - cos^2\theta_+ & 0 & 0 \\ cos^2\theta_- - cos^2\theta_+ & cos^2\theta_- + cos^2\theta_+ & 0 & 0 \\ 0 & 0 & -2cos\theta_+ cos\theta_- & 0 \\ 0 & 0 & 0 & -2cos\theta_+ cos\theta_- \end{bmatrix} \begin{bmatrix} 1 \\ 0 \\ 0 \\ 0 \end{bmatrix} \qquad \text{Eq. 2}$$

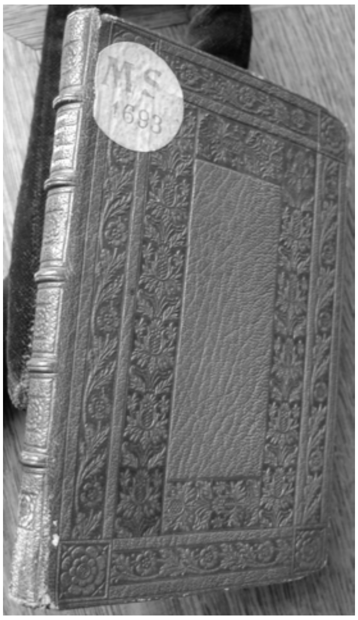

Figure 2. *L'Agenda de Malus*. Photograph by the author. *Bibliothèque de L'Institut de France, Paris.*

In 1798, Bonaparte wanted a colony to compete with British India and tried to conquer Egypt. But, he was keen to capture the culture as well as the country. Malus was one of the 151 scholars comprising the so-called Commission on Arts and Sciences that Bonaparte brought with his army. Malus was also a founding member of the Institute of Egypt,[7,8] an elite subgroup of commissioners, along with some of the greatest scholars of his time including the mathematician Joseph Fourier, the chemist Claude Louis Berthollet, and the biologist Étienne Geoffroy Saint-Hilaire. Malus was the protegé of the mathematician Gaspard Monge (Figure 1), Napoleon's close companion throughout the Egyptian compaign. Malus disembarked with the army near Alexandria and marched towards Cairo.

As practitioners of polarized light spectroscopy, polarimetry, and ellipsometry, we could not help but admire the contributions of Malus to science. However, our interest in Malus was heightened in the Spring of 2003 when the American President, George W. Bush, announced his invasion of Iraq. Bush's declaration of war was practically plagiarized from a justification for the invasion of Egypt by Bonaparte 205 years earlier. Below I compare three paragraphs, first Bush, followed by Bonaparte in *italics*.[9]

> My fellow citizens, at this hour American and coalition forces are in the early stages of military operations to disarm Iraq, to free its people and to defend the world from grave danger.[10]
>
> *In the name of the French Republic, founded on liberty and equality, the commander-in-chief of the French armies, Bonaparte lets it be known to the whole population of Egypt that the beys who govern Egypt have insulted the French nation and oppressed French merchants long enough: the hour of their punishment has come.*
>
> We come to Iraq with respect for its citizens, for their great civilization and for the religious faiths they practice. We have no ambition in Iraq, except to remove a threat and restore control of that country to its own people.
>
> *Peoples of Egypt, you will be told that I have come to destroy your religion. Do not believe it! I have come to restore to you your rights and to punish the usurpers. I respect His prophet Mohammed and the admirable Koran.*
>
> Now that conflict has come, the only way to limit its duration is to apply decisive force. And I assure you, this will not be a campaign of half measures and we will accept no outcome but victory.
>
> *But woe, woe to those who side with the Mamelukes and help them to make war on us. There shall be no salvation for them, and their memory shall be wiped out.* (translation by Herrold[9])

In the words of the songwriter Elvis Costello, "History repeats the old conceits/The glib replies the same defeats."[11] Both Bonaparte and Bush, predictably, met fierce, guerilla insurgencies. “Why, after the defeat of the Mamelukes [Ottoman overlords] and the taking of Cairo,” asked historian Charles-Roux, “did the French continue to experience the hostility of the natives in many regions? Because after…they remained, in the eyes of the Mussulmans, infidels whose intrusion into the territory of Islam was a profanation…On this account they inspired an antipathy, a repulsion, which could breed hatred…”.[12]

We wondered to what extent we could learn something about our national catastrophe by walking in the steps of Malus who was in the thick of the French invasion. We knew that Malus kept a diary while in Egypt. It was published in 1892,[13] but we sought out the original at the Institute of France in Paris (Figure 2). Here, is Malus writing of the siege of Jaffa:

> …soldiers scattered throughout slitting the throats of men, women, elderly, children, Christians, Turks; all that bore the human form was victim of their fury. The tumult of carnage, the broken doors, the houses shaken by the noise of gun shots, the cries of the women, the father and child overthrown one on the other, the violated daughter on the corpse of her mother; the smoke of dead bodies burned in their garments, the smell of blood, the groans of the wounded, the cries of the conquerors disputing over the spoils of their expiring prey, infuriated soldiers responding to the cries of despair with exclamations of rage and redoubled blows. Lastly, men satiated with blood and gold, falling down in mere weariness on the heaps of corpses—such was the spectacle that this unfortunate city presented until night.[13]

Malus was the conscience of the army.

Following this massacre, there was an outbreak of the plague. Malus fell victim and waited for his demise. But miraculously, he recovered. While regaining his strength in the desert, he wrote a memoir on the nature of light.[14] He imagined that light was an admixture of oxygen and caloric, with colors determined by the proportions of these two substances. The work, derived from lessons that Malus likely received as a student at *École Polytechnique*, was off the mark. But, nevertheless, Jean-Baptiste Biot (see section 3) captured something of Malus' character when he remarked that "No army in the world ever before counted in its ranks an officer who occupied himself in the spare hours of advanced posts with researches so complete and so profound."[4]

Malus barely made it back Paris to restart his scientific studies, culminating in his discovery of polarization by reflection. Malus adopted the position that light particles carried with them some dissymmetry directed at right angles to the direction of propagation, but oriented every which way in direct sunlight. Rays with directed dissymmetries were selected from a random bundle of rays during the process of reflection. [15] This Newtonian, particle-like conception of light was likewise still prejudiced by his schooling.

But, the wave theory was cresting. Indeed, it was Thomas Young who wrote to Malus with the good news that the Frenchmen was to be awarded the Rumford Medal of the Royal Society of London (1811). Malus died the next year, most likely from tuberculosis, his health forever compromised since his bout with the plague. Yet, a great tradition of research on and with polarized light in France was set in motion by his discovery.

3. François Arago (1786-1853)[16]

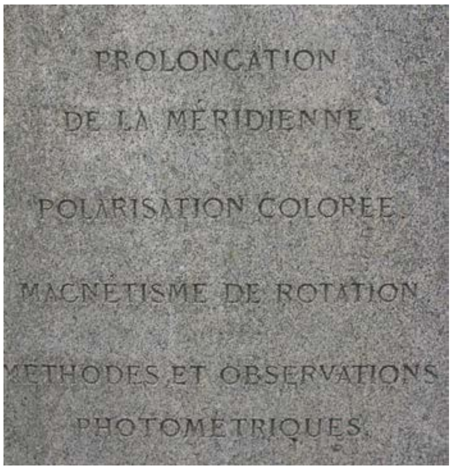

FIGURE 3. Achievements of Arago etched on his tombstone, Père Lachaise cemetery. *Polarisation colorée* was a term introduced in a paper describing both the chromatic phenomena from thin mica plates between crossed polarizers (interference due to phase differences in the recombined rays traversing the anisotropic plate) and optical rotatory dispersion in quartz. We now understand one is a manifestation of linear birefringence and the other a manifestation of circular birefringence. Photograph by the author.

Only on his deathbed in 1853 did François Arago (see Figure 1) completed a biographical sketch of his friend Malus, more than 40 years after Malus died.[17] Recording the life of Malus was something that remained for Arago a job long undone for decades and became an urgency when Arago was running out of time. Arago admired Malus for his science -- Arago championed the appointment of Malus to the Academy of Sciences -- and for his character. Of Malus' account of the massacre at Jaffa, Arago wrote: “This forcible passage…is the faithful picture of what happens in every town taken by storm…which the narrative of Malus has revealed in all their horrors. Our condemnation will be reserved for those who provoke these impious wars, which have no other motive than personal ambition, and the desire for a vain and false glory."[4]

When Arago himself was young, the National Assembly decreed that the meter was to be measured as one ten millionth of the 90° arc of latitude from the North Pole to the equator through Paris.[18,19] They sent Arago and another promising young scientist, Biot (see section 3), on a mission to measure the shape of the Earth by triangulation. Arago and Biot left Paris, heading South. When they reached Spain, Biot returned to Paris with preliminary data. Arago continued on, lighting beacons on mountaintops in Majorca. However, this was during the brutal Peninsular War (1808-1814) between France and Spain. The Spaniards were suspicious of a Frenchman lighting beacons on mountaintops. Arago was imprisoned at gunpoint, escaped, was recaptured twice and escaped twice more. After two years wandering around North Africa, Arago returned to Paris, long having been given up for dead. He was welcomed as a hero in the service of the Republic and quickly elected to the French Academy of Sciences.

Arago did not want to be an *Académicien* because he survived an ordeal. He wanted to be recognized for making a signficant contribution to science. It did not take him long to do so. Arago passed white light, polarized by reflection according to Malus, along the high symmetry axis of quartz. When viewed with an analyzer (a second pile of polarizing plates), Arago witnessed the dispersion of colors depending on the relative orientations of polarizer and analyzer. Arago called this phenomenon *polarisation colorée* (Figure 3).[20,21] We know it as optical rotatory dispersion (ORD). although his understanding of the process was confused.[15] In the same *Mémoire*, Arago also described thin film interference colors produced by sheets of mica between crossed polarizers. *Polarisation colorée* is best taken literally: "colors made with polarized light". Longchambon wrote, "Arago did not distinguish sharply between rotatory polarization and chromatic polarisation. It is to Biot that we owe a complete study of these two phenomena..." (translation of Lowry[22]). In his biography of Arago, Lequeux distinguishes the two phenomenological observations as chromatic polarization (a manifestation of linear birefringence) and *rotational chromatic polarization* (a manifestation of circular birefringence).[23] Rotational chromatic polarization, or ORD, is expressed for any one color as a rotation matrix applied to linearly polarized light as in Eq. 3.

$$\begin{bmatrix} 1 \\ cos2\theta \\ -sin2\theta \\ 0 \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & cos2\theta & sin2\theta & 0 \\ 0 & -sin2\theta & cos2\theta & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 1 \\ 1 \\ 0 \\ 0 \end{bmatrix}. \qquad \text{Eq. 3}$$

Arago recognized that interference colors from mica could be used as a sensitive indicator of polarized light. He illuminated mica between polarizers with moonlight and in this way established that light from the moon was sided by reflection.[24,25]

By 1815, Arago was famous. That year, Bonaparte, now Emperor Napoleon, was defeated at Waterloo, and recognizing an end to his military career, he turned to science. Napoleon explained to his friend Monge,

> Since I would no more be allowed to command armies, I see that only science would be able to take strongly possession of my soul and of my mind. To learn [what] others have done would not be enough for me. I want, in this new career, to do works and make discoveries worthy of me. I need a companion who would first instruct me rapidly of the present status of science. Then we would travel together through the New World...during this immense trip, we would study all the great phenomena of the physics of the Earth that the scientific world has not yet described.[23]

Monge volunteered for this mission but Napoleon suggested that the famous geometer was too old. He proposed Arago instead.

4. Jean-Baptiste Biot (1774-1862)

Jean-Baptise Biot (Figure 4) was jealous of *la gloire* earned by Arago, his partner in triangulation. Biot quickly moved to clarify the difference between quartz and mica with respect to *polarisation colorée*.[26] Then, he made an enduring discovery, that a variety of naturally occurring compounds also have the capacity to rotate the plane of polarization when in solution.[27]

Arago felt that Biot had intruded upon his scientific territory. A rift developed that widened throughout the lives of the two physicists. Nevertheless, Biot persisted in his researches into optical activity, establishing that first dispersion relations for optical activity and the inverse square dependence on wave length.[28]

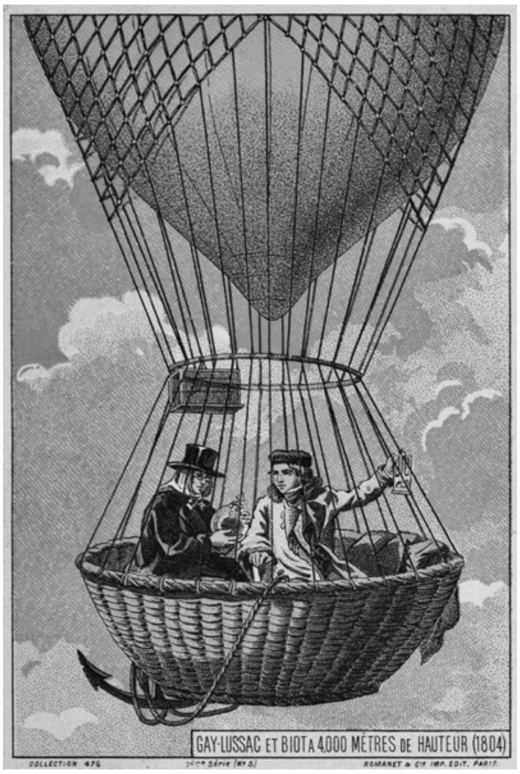

FIGURE 4. Biot (right) and Joseph Gay-Lussac in a balloon. *Gay-Lussac et Biot à 4000 mètres de Hauteur* (1804). Paris : Romanet & cie., imp. edit., [between 1890 and 1900]. Wikimedia Commons.

In the 1830s, Biot improved the sensitivity of the optical polarimeter.[29] Most organic compounds Biot could isolate now seemed endowed with optical activity and he came to see the influence of dissolved substances on polarized light as a signature of a vital force separating animate and inanimate matter that only he could detect. According to Levitt, in her admirable dual biography of Arago and Biot, "Optical activity, Biot insisted, was not a property of organizational structure. It was, rather, the sole means in mans's possession of confronting the otherwise undefinable limit between light and nonlife on the molecular level."

As they aged, Arago and Biot argued not only about optical activity but also the discovery of photography, politics -- Arago was a Republican and Biot a Royalist -- and most significantly, they argued about slavery. Arago was an abolitionist while Biot favored the Atlantic slave trade that supported the lucrative French colonies in the West Indies.[24]

In fact, Biot aimed to place the entire French colonial economy on the foundation of polarimetry. The precise quantity of sugar in a batch of molasses distilled from cane sugar was uncertain, but sucrose was what customers were ultimately buying. Biot was keen to quantity this commodity with polarized light and towards this end he developed a polarimetric technique especially adapted to molasses that he called saccharimetry.[30, 31] Fortunately, his program was never fully advanced. Following the abdication of King Louis Philippe in 1848, Arago became the minister of the colonies in the provisional government. Within two weeks of assuming this post, he abolished slavery throughout the French empire:[32,33]

> The provisional government, Considering that slavery is an assault against human dignity; That by destroying the free will of man, it suppresses the natural principle of right and duty; That it is a flagrant violation of the republican dogma: Liberty, Equality, Fraternity[34] (translation, Levitt[24]).

The political pendulum swung back in Biot's favor when Louis Napoléon declared himself Emperor Napoléon III in 1852. Arago was dismissed of his official duties. The next year, Arago was dying. He was blind and infirmed as a consequence of diabetes, a disease Biot had proposed to monitor with the saccharimeter.[35] Biot came to visit Arago, under the pretext of delivering Arago's salary as Secretary of the Academy of Sciences, uncollected for months because Arago's physical deterioration prevented him from attending to Academy business. As Levitt explains, this was one last disagreement between the two rivals. Arago refused to accept compensation for a job that he was unable to perform.[24]

5. Augustin-Jean Fresnel (1788-1827)

Arago championed the investigations of Augustin-Jean Frensel (also named on the Southeast side of the Eiffel Tower) on the diffraction of light.[36] These seminal studies established the wave theory of light by confirming Thomas Young's[37] observations of diffraction and the transverse nature of the oscillations, results that Fresnel came to independently and put on a stronger quantitative foundation.

In 1817, Fresnel discovered circularly polarized light (CPL) produced by a prism of his own invention. Rather than relying on the anisotropy of crystal to produce phase shifts between eigenpolarizations in crystals, Fresnel's rhomb exploits two total internal reflections.[38] The refractivity of the glass and the angles of two reflections inside the prism are tuned so that a phase shift of $\pi/4$ is produced between the *s* and *p* components of the incident light at each of the two interfaces, reflections 1 and 2. The Mueller matrix (Eq. 4) is that for a quarter wave plate:

$$M_{\frac{\pi}{2}} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & \cos(\theta_1+\theta_2) & -\sin(\theta_1+\theta_2) \\ 0 & 0 & \sin(\theta_1+\theta_2) & \cos(\theta_1+\theta_2) \end{bmatrix}$$

Eq. 4

Light linearly polarized at +45° $[1,0,1,0]^T$ is transformed to right circularly polarized light, $[1,0,0,1]^T$.

Arago urged Fresnel to investigate the interference fringes between the two rays emerging from a birefringent crystal. There seemed to be no effect of one upon the other. In this way, Fresnel articulated the laws that govern the interference of polarized rays: incoherent light does not interfere whereas coherent light only interferes for parallel polarizations.[39] A modern perspective on the equivalence of Young's double-slit and crystalline double-refraction interference experiments was recently reported, a demonstration made with Mueller matrix polarimetry.[40]

Fresnel, before his contributions to the science of light, was a young graduate of *École nationale des ponts et chaussées* stationed in the south of France when Napoleon I returned from Elba and attempted to march on Paris. Fresnel joined the army raised to block Napoleon's path to the capital. Fresnel's actions were seen by some as traitorous, but he was a good engineer and merely furloughed to his hometown of Mathieu as punishment. During this time, Fresnel completed his major work on diffraction.[38]

Napoleon's return was short (100 days), ending in the defeat at Waterloo. Fresnel was reinstated and posted to Rennes in 1816. Of the nine scientists portrayed in this article, Fresnel is the only one, to the best of our knowledge, to have lived in Rennes, the city hosting the 16th International Conference on Chiroptical Spectroscopy, June 2017.

1816 was the so-called Year There Was No Summer[41] because of climate abnormalities associated with the huge eruption of Mount Tambora in the Dutch East Indies (Figure 5). It snowed in France in June. There were worldwide crop failures. According to the historian Levitt, Brittany "was particular hard hit.

FIGURE 5. Eruption of Mt. Tambora, Dutch East Indies, 1815, the cause of The Year with No Summer. Public domain: https://www.treehugger.com/energy-disasters/200-years-ago-today-mount-tambora-exploded-and-changed-world.html.

The streets of Rennes filled with men, women, and children, 'all pale with hunger', starved out of the countryside." These poor souls were conscripted for public works projects supervised by Fresnel. Fresnel is widely described as a sensitive soul. He did not enjoy corralling the hungry and destitute to serve as laborers. "I burn to leave Rennes," he wrote to Arago.[42]

During the same week in 1819 that Théodore Géricault showed his painting of *The Raft of the Medusa*, a shipwreck, Augustin-Jean Fresnel filed his patent for the so-called Fresnel lens, a device that saved countless lives by increasing the effectiveness of lighthouses.[42] Fresnel realized that the magic of glass lenses all happens at surfaces. The bulk of the glass served no optical purpose and was merely dead weight. By "flattening" a lens and making it mostly surface in concentric inclined rings, giant, lenses with short focal lengths could be installed in lighthouses, broadcasting strong beams to points far from the shore.

Fresnel, during his lifetime, received *plus de gloire* from his lens than for any of his scientific achievements. The importance of lighthouses for navigation are now much diminished but those of a certain age can now carry plastic Fresnel lenses in our wallets in order to read the bistro menu in candle light.

In 1823, Fresnel derived his reflection and transmission laws for plane waves striking the interface between two isotropic, dielectric media (e.g. air and glass).[43] In our work, reckoning the balance between reflection and transmission of light striking a non-normal interface of a medium that is both anisotropic and optically active has been critical.[44,45,46,47,48,49]

In crystal optics, accounting for the incoherent superposition that may arise from multiple reflections in thin layers that need not be coherent requires that the optical transfer matrices are first converted to coherency ($\mathcal{C}$) or second moment matrices before summation to capture cross correlations between polarization components. $\mathcal{C} = \mathbf{J}\otimes\mathbf{J}^*$ [50] where $\mathbf{J}$ is a Jones matrix. The corresponding Mueller matrix can be calculated by $\mathbf{M}=\mathbf{A}(\mathbf{J}\otimes\mathbf{J}^*)\mathbf{A}^{-1}$, where A = [1,0,0,1; 1,0,0,-1; 0,1,1,0; 0,*i*,-*i*,0], and $\otimes$ is the Kronecker product.

At issue is precisely that same kind of question that first consumed Arago and Fresnel: How do polarized lights interfere?

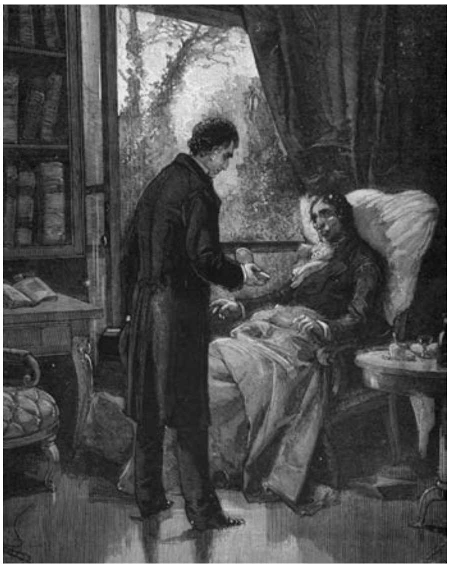

FIGURE 6. Arago bringing a dying Fresnel the Rumford Medal (*La Science Populaire. Journal Hebdomadaire Illustré*, August 19, 1880). Arago wrote to Thomas Young that Fresnel was "half dead" when he delivered the medal.[23]

As did Malus, Fresnel was awarded the Rumford medal (1824). As did Malus, Fresnel died young, age 39 (Figure 6).

6. Louis Pasteur (1822-1895)

The reader, like the author, may be tired of Pasteur and his tartrate crystals (Figure 7).[51] So much has been written about them. Pasteur's separation of enantiomorphs is now lauded as much for its popularity as for its ingenuity. It has become a celebrity experiment.[52,53,54]

Less well known is Pasteur's homage to Fresnel,[55] recorded by Lowry,[22] conceding that Fresnel had early and excellent intuition about the cause of rotatory polarization. In 1825, Fresnel wrote, "Rock-crystal shows optical phenomena which

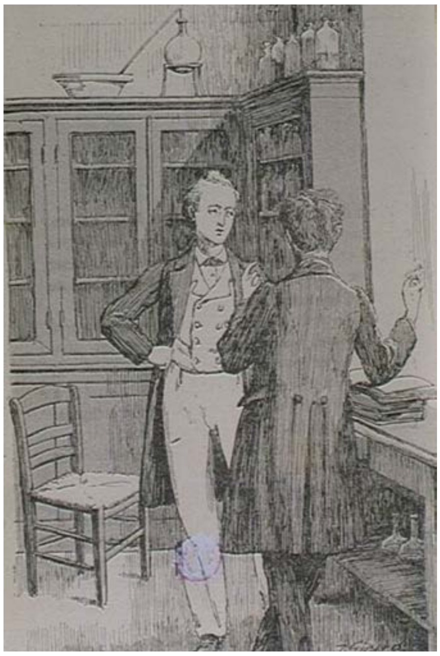

FIGURE 7. Louis Pasteur explaining to Claude Bernard his discoveries in crystallography and molecular dissymmetry. *Louis Pasteur explique à Claude Bernard ses découvertes sur la cristallographie et la dissymétrie moléculaire, vers* 1848/1850 (after J. Girard, 1887). [Service photo IP] [Cote: D1595]. gallica.bnf.fr/Bibliothèque nationale de France. Pasteur's back is towards us, the figure that is explaining. The facing figure resembles Bernard.

cannot be reconciled with complete parallelism of the molecular lines, and which would seem to indicate a progressive and regular deviation of these lines in the passage from one layer of the medium to the next."[56] Here, Fresnel foreshadows Pasteur's discovery, while making a definite statement about the arrangements of atoms in space.

One unfortunate consequence of the love of all things Pasteur by the community of chemists is that the necessary condition for optical activity has been persistently misstated. The idea that chirality is a necessary condition for optical activity is inherited from Pasteur, but this only applies to isotropic media in which molecules are randomly reorienting. Gibbs[57] first recognized that some achiral crystals could indeed be optically active, and this was stated formally by Voigt.[58] However, the verification of Gibbs' prediction had to wait for an elegant experiment by Hobden[59] on achiral crystals of $AgGaS_2$ with which optical activity was measured in a low symmetry direction of this achiral crystal at a particular frequency at which the refractive indices of eigenmodes in the crystal cross and the linear birefringence drops to zero, leaving only circular birefringence. He said, “It is hoped that this observation will finally eradicate the notion that optical activity is exclusively related to enantiomorphism.” It didn’t. Hansen and Bak[60] computed the rotatory strength of the HOMO-LUMO transition of *cis*-butadiene with C_{2v} symmetry and reiterated “that certain classes of achiral molecules may exhibit optical rotatory power under anisotropic conditions" while conceding that this "apparently has not attracted much attention in the community of structural chemists, although the phenomenon is part of the general theory of crystal optics.” The chiroptical tensor elements are given routinely for achiral point groups D_{2d}, S_4, C_{2v} and C_s in crystal physics texts.[61] However, our intuition is based on experience, and since generations of chemists only observed optical activity with chiral analytes, our incorrect intuition was reinforced. This is a prejudice associated with the fact that it is quite difficult to measure optical activity in organized media.[62,63,64]

The story of the discovery of the link between macroscopic and microscopic chirality that Pasteur tells so romantically in 1860[55] fails to mention that along with old Biot who insisted that Pasteur perform his improbable resolution at the *Collège de France* under the senior colleague's watchful eye, there was another scientist in the room, Henri de Sénarmont.[65,66] We had earlier studied Sénarmont's career because of an experiment that he carried out in 1854. He wanted to understand why some minerals like tourmaline were dichroic when reoriented in polarized light. Since he could not prepare tourmaline in the laboratory, he tried to make *faux* tourmaline by precipitating a salt in the presence of a textile dye. He prepared dichroic strontium nitrate tetrahydrate stained with the extract of logwood bark, rich in a red quinone.[67,68] This was the first purposeful instance of the dyeing of a growing crystal, a subject that we have analyzed at length over the years.[69,70] We have long aspired to measure artificial circular dichroism in dyed crystals, but this has been a much harder proposition than the measurement of artificial linear dichroism.[71]

Nevertheless, Biot found in Pasteur a like-minded champion of vitalism. They both believed that the action of molecules on polarized light was a signature of life. According to Levitt:

> Biot and Pasteur made clear the enemy they sought to vanquish, materialism. But the materialists saw themselves as the heroes of their own drama, fighting against the dark forces of superstition. The vitalists saw the denial of the distinction between life and nonlife as the denial of whatever was transcendent in man. The materialists saw the intervention of spirit in the discussion of forces as dangerous incursion into human autonomy. In both cases, the crucial implication was that of human freedom.

7. Frédéric Wallerant (1858-1936)[72]

In the previous section, we in no way meant to undercut the significance of Pasteur's great insight, seeing microscopic molecular configuration manifest in macroscopic crystalline form. Unfortunately, conglomerates that deposit well-formed crystals with hemihedral facets that can be mechanically separated are few and far between.[73] Frédéric Wallerant was keen to find other ways of making configuration manifest in crystal form.

Wallerant seized upon the observations of Michel-Lévy (with Munier-Chalmas) -- best remembered for the eponymous color chart for the characterization of the birefringence of minerals in terms of the Newton's interference colors -- that chalcedony, a fibrous form of quartz, sometimes grows as helicoids as evidenced by periodic extinction bands between crossed polarizers (Figure 8).[74] This observation, combined with the arrival of cholesteric organic liquid crystals in Paris[75,76] that also showed rhythmic bands of optical contrast as a result of the precession of anisotropic bodies, pushed Wallerant to attempt to grow helicoidal crystals in the laboratory. He aimed to direct the sense of the helicoids by inducing twisting with chiral dopants.

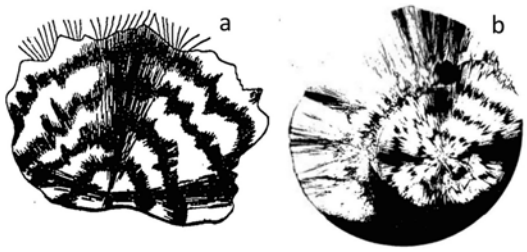

FIGURE 8. Sketch (a) and photograph (b) of banded spherulites of chalcedony from ref. 74.

Achiral crystalline compounds such as malonamide and glycolic acid, when grown from the melt to which he added chiral compounds, gave optical signatures that resembled chalcedony.[77,78] The sense of twist could be established by rotating the helicoids about their long axes, and watching the periodic extinction bands move up or down like a rotating barber-shop pole. Indeed, the sense of twist could be well correlated with the configuration of the additives such as (±)-tartaric acids.[79]

Wallerant thought he had made a discovery of Pasteurian significance. “The twisting,” he said, “constitutes, with the

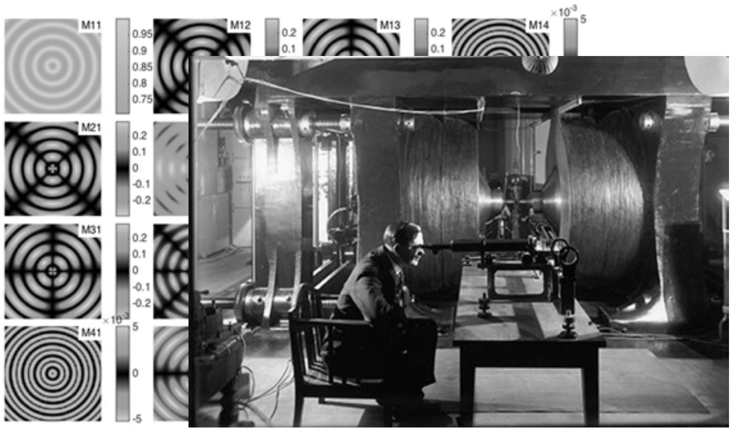

FIGURE 9. The simulated 4×4 Mueller matrix elements of twisted, banded spherulites with dye aligning perpendicular to the fiber growth direction. The unit is radians for all the elements.

FIGURE 10. Spectroscope aimed at the giant electromagnet built under the direction of Aimé Cotton in 1928. Fonds historique/CNRS PHOTOTHEQUE.

dissymmetry of the crystalline form discovered by Pasteur, the only known manifestation in materials of the dissymmetry of active substances."[78] And in a later paper: "Thus far we know of three kinds of anisotropic structures: crystals, the anisotropic drops of Lehmann [liquid crystals], and twisted crystals."[79]

We repeated Wallerant's experiments on the twisting of rescorcinol induced by (±)-tartaric acids.[80,81] This work, combined with our studies[80,81,82,83,84,85,86] of the extraordinarily common but wholly unappreciated phenomenon of helicoidal crystal growth, has convinced us that Wallerant did indeed make some very important discoveries but they failed to attract any attention for more than a century.

Of course, if you can dye crystals, you can dye twisted crystals. By doing so, we made spherulitic crystal aggregates whose linear dichroism manifests as a helicoidal progression with absorbing chromophores that spiral around and within twisted crystals (Figure 9).[87] Here, the material is not homogeneous along the light path but rather traverses many misoriented crystallites. The overall polarization transformation matrix must be constructed from products of Mueller matrices for single crystals, multiplied by rotation matrices that represent the arrangement of components. A simulated Mueller matrix micrograph, well matched to data, is displayed in Figure 9.

As with so many other scientists, the research of Wallerant was transformed by the discovery of X-ray diffraction. With his assistant Charles Mauguin, who deftly used polarized light to study liquid crystals, Wallerant set up X-ray equipment at the Sorbonne in 1919. This, lab, along with that of Charles Friedel at *École des Mines*, became the leading centers physical crystallography in France.[88,89]

8. Aimé Cotton (1869-1951)

Lowry[22] ascribes the discovery of circular dichroism (CD) to Haidinger[90] and Dove[91] who investigated the differential absorption of left and right circularly polarized light in amethyst, the purple form of quartz. This observation was challenged by Perucca,[92] as described in our papers,[71,93] who insisted that the anomalous biaxiality of amethyst contributed a parasitic linear anisotropy to the early CD measurements, thus producing artefactual CD signals. Recently, *bona fide* CD was demonstrated in uniaxial synthetic amethyst.[94] As such, there remains some uncertainty as to what was measured by Haidinger and Dove, given that the pitfalls of measuring CD in anisotropic crystals were not appreciated in the middle of the 19th Century.

This may leave Aimé Cotton (Figure 10) as the proper discoverer of CD by experiment. He measured the CD of a solutions of potassium chromium tartrate and potassium copper tartrate and thus was not burdened by artifacts of measurement arising from the mixing of polarization states and imperfectly polarized inputs. (±)-Tartrates gave equal and opposite spectra while racemates were inactive.[95,96,97,98] Irrespective of the amethyst mystery, Cotton's name is now synonymous with CD; he coined the term, and peaks in CD spectra are universally known as Cotton effects.[99]

Cotton made his measurement as a graduate student working at *École normale supérieure,* where he studied and worked alongside Paul Langevin, Jean Perrin, Pierre Weiss, and Pierre Curie. His discovery originated in the observation of anomalous ORD near absorption bands.[100] He reasoned:

> In an optically-active medium, there are also [in addition to anisotropic media] two kinds of rays, a right circularly polarised ray and a left circularly polarised ray, which are propagated with different velocities, and it is seen, in some cases, that a ray is sharply separated, in traversing the medium, into two circularly polarised rays of oppoiste sign.
>
> If this circular double refraction is compared with crytsalline double refraction, a very close analogy is noticed between the two types of phenomenon. It is therefore natural to ask if there are not active substances which absorb unequally a left and a right ray".[96] [Translation, Lowry[22]]

Cotton's contribution depended on Fresnel's achromatic rhomb for transforming linearly polarized light into CPL by successive reflections. Cotton placed two such rhombs side by side with their principal sections oriented at 90° with respect to one another, so that he could compare the transmission of left and right CPL directly.

After the discovery of CD, Cotton worked especially closely with Weiss. Together they studied the Zeeman effect, and made an accurate measurement of the charge to mass ratio of the electron. During the First World War, they developed the Cotton-Weiss system for locating enemy artillery. From the time differences in signals recorded at remotely separated sound receiving stations, the positions of guns were computed.

In 1913, Cotton married Eugénie Feytis, herself a physicist having trained with Marie Curie. Eugénie was later director of the *École normale supérieure de Sèvres* but she was fired by the Vichy government because she was viewed as both a communist and a feminist. Indeed, she was a founder of the *Union des femmes françaises,* and the Women's International Democratic Federation where she served as president for more than 20 years. She advocated for the decolonization of Algeria and Vietnam and provided refuge to Spanish and German anti-fascists.

Aimé was likewise described as a profound humanist. He was a champion of the international language Esperanto which he hoped would lead to a greater understanding among nations. He worked for the resistance and was twice imprisoned. In 1941, Cotton was held at the notorious Fresnes prison where the SS tortured members of the French resistance. Charles Mauguin wrote to Eugénie after Aimé's death "Naturally benevolent, [Amié Cotton] gave himself unreservedly with unflinching dedication, to the cause he felt was right, but took a courageous and sharp stand against what he disliked. This is undoubtedly what got him, in 1941, to be interned by the Germans in Fresnes, where we were both, each without the knowledge of the other. I will never forget our return together on the metro, leaving prison with the shaggy heads of highway robbers." Eugénie met her husband at the Montparnasse Station, "an old man, exhausted and emaciated, dragging shoes without laces, neither ties nor suspenders, and carrying under his arm a poor little bundle wrapped in his dressing-gown and tied with the sleeves."[1]

Aimé was released but rearrested in 1942, along with his son Eugene because of the latter's work with the resistance in Lyon. Aimé was later awarded the *Rosette de la résistance*.

9. Francis Perrin (1870-1942)

Francis Perrin was the son of Jean Perrin, Cotton's companion, whose studies of Brownian motion confirmed the atomic nature of matter[101] and realized the predictions of Einstein. Francis was raised within the rich scientific culture of his father's inner circle including Marie Curie and Paul Langevin.[102] In a postscript to Einstein, Jean crows about Francis' performance in school.[103] Imagine bragging to Einstein that your kid is smart. Francis must have been way ahead of the pack.

Rotational diffusion assayed by fluorescence depolarization, the subject of Francis' PhD thesis in physics,[104] was a subject the younger Perrin was born to. In fact, Francis also earned a PhD in mathematics on Brownian motion,[105] the latter of which was supervised by Émile Borel. Borel was imprisoned by the Vichy authorities along with Cotton. In 1926, Perrin *fils* wrote the well-known equation expressing fluorescence polarization as a function of the size of the emitter, the lifetime of the excited state, temperature, and solvent viscosity.[106,107] He also first reported depolarization as a consequence of resonance energy transfer.[104]

In 1942, Perrin wrote out the transformation of one Stokes vector to another in terms of 16 parameters in four linear equations.[108] Hans Mueller placed these coefficients in the form of a matrix.[109] [110] Only in this form did Perrin's conception -- predated by another Frenchman, Soleillet[111,112] -- get put to practice. Thus, the polarization transfer matrix is known as the Mueller matrix, aided somewhat by alliteration, and not the Perrin matrix, despite the fact that Mueller gave priority to Perrin.[113]

Here, we propose calling the Mueller matrix for fluorescence, the Perrin matrix (**P**). That would be fitting. $\mathbf{P} = \mathbf{M_1 S M_0}$, where $\mathbf{M_0}$ describes the absorption and retardation effects that the ground molecular state induces on the incident (excitation) beam, $\mathbf{M_1}$ accounts for the effects of the medium on the emitted light, and **S** is scattering matrix that captures the polarization transformation between the ground and the excited state, either by transformation of ground and excited state dipoles, or rotational diffusion.[114] In principle, formulation of fluorescence in this way should yield all the fluorescence polarization parameters including fluorescence anisotropy (FA, Eq. 5), fluorescence detected circular dichroism (FDCD, Eq. 6), and circularly polarized luminescence (CPL, Eq. 7), albeit only in the absence of bulk anisotropy of the medium:

$$FA = \frac{P_{22}-P_{21}}{3(P_{11}-P_{12})+(P_{21}-P_{22})}, \quad \text{Eq. 5}$$

$$FDCD = \frac{P_{14}}{P_{11}}, \text{ and} \quad \text{Eq. 6}$$

$$CPL = \frac{P_{41}}{P_{11}}. \quad \text{Eq. 7}$$

Perrin turned his attention increasingly to nuclear physics as both he and physics matured. He served as the high commissioner (Figure 11) of the French atomic energy agency (1951 - 1970), replacing Frédéric Joliot-Curie because the latter was opposed to weapons research (Alternatively, Joliot-Curie was sacked because of Stalinist tendencies).[115] Perrin was instrumental in the development of France's nuclear weapons program,[116] and as he revealed in the *Sunday Times*, in 1986, France was also instrumental in helping Israel build nuclear facilities in Dimona: "We considered we could give Israel the secrets provided they kept it to themselves."[117, 118] Perrin was a forceful advocate for the peaceful use of atomic energy, a legacy preserved in the comparatively high percentage of nuclear energy in France's electricity generation portfolio. Perrin initially served de Gaulle but joined leftist protests in 1958.


Administration-Rédaction : 142, rue Montmartre, Paris-2e
SEMAINE DU 16 AU 22 AVRIL 1950

Pour remplacer Joliot l'atomique Zoé hésite entre six prétendants

« JAMAIS un véritable savant progressiste ne donnera une parcelle de sa science pour faire la guerre à l'U.R.S.S. »

Ces déclarations fracassantes de Joliot-Curie, au Congrès communiste de Gennevilliers, attirent de nouveau l'attention sur ce paradoxe. La France tolérera-t-elle plus longtemps que le maître de ses recherches atomiques se réclame d'un parti qui n'est plus français ?

Que M. Joliot-Curie choisisse entre son poste et le parti communiste, tel est le dilemme. Et puisqu'il s'obstine à ne pas vouloir le trancher lui-même, le gouvernement devra le trancher à sa place.

La succession de Joliot-Curie va donc s'ouvrir.

Bertrand GOLDSCHMIDT

Francis PERRIN

Jean THIBAUD

Louis DE BROGLIE

FIGURE 11. *La Presse*, 16-22 April 1950. Bibliothèque nationale de France.

The character of the Perrins, both *père* and *fils*, was nicely summed up by Beraban-Santos. Both Jean and Francis, were socialists and atheists. Like many nineteenth century [scientists], Jean Perrin viewed science almost as a religion. In the words of Raspail, inscribed in the surviving pedestal of his statue, located near Francis Perrin's residence...*la Science, l'unique religion de l'avenir* (Science, the only Religion of the future). Francis Perrin publicly denied all religions and gods, and was the President of Honour of the Union des Athés. He donated his body to science.

Jean and Francis Perrin were, in their own way and in the best French spirit, defenders of the *Droits de l'Homme*, and rejected all totalitarianisms. In the difficult period between wars, both actively opposed the rising fascism in Europe.[102]

10. Alain Aspect (1947-)

Malus' discovery of the polarization of light by reflection led to the development of the wave theory of light and the science of molecular chirality in the 19th Century, and more. In the 21st century, the strangest phenomena and the most fantastic technologies—nonlocality and quantum computing, respectively—rely on entangled, polarized photons. Entangled particles behave in the same way when measured along the same axis.

Malus meets quantum mechanics in the work Alain Aspect,

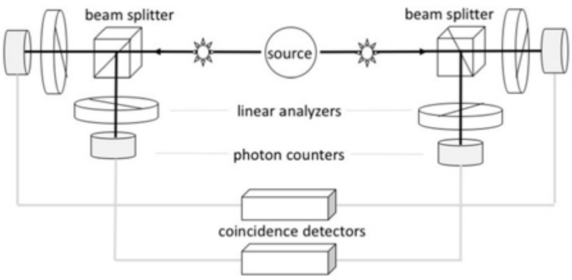

FIGURE 12. Scheme of so-called Bell/EPR experiment of the kind performed by Aspect and coworkers. A source generates a pair of entangled photons. (Today this is typically achieved by parametric down conversion from an optically nonlinear crystal). The photons are launched towards beam splitters that direct the photons to one of two polarizer/detector pairs at each end of the apparatus. Photon counting detectors on opposite sides are connected by coincidence detectors. The key in the Aspect experiments is the fact that the state of the polarizers was not established until after the photons pair was created.

the Augustin Fresnel Professor at *Institut d'Optique, Ecole Polytechnique*. Biographical accounts of Aspect invariably emphasize his national service for three years (1972-1974) in Cameroon, during which he studied the new quantum mechanics textbook of Claude Cohen-Tannoudji *et al*.[119] in order to fill gaps in his training that emphasized classical physics. Aspect appreciated this book because it was "...neutral with respect to the foundations [of quantum mechanics]. No brainwashing...".[120] As a PhD student at the *Université d'Orsay* from 1975-1983, unburdened by prejudice, Aspect decided to use polarized, entangled photons to put the philosophical foundations of quantum mechanics to the test.[121,122]

It is well known that Einstein was uncomfortable with quantum mechanics and was keen to find its soft underbelly. His debates with Bohr have generated volumes of commentary.[123,124] Bohr seemed to have had the upper hand, until 1935 when Einstein published his so-called EPR paradox, named for collaboration with Boris Podolsky and Nathan Rosen. Einstein and his younger colleagues asked, "Can quantum-mechanical description of physical reality be considered complete?"[125] They proposed a thought experiment: Imagine the generation of a pair of particles, say polarized photons, in correlated or entangled polarization states flying to opposite sides of the universe to conserve momentum. If you should measure the particle on the right side of the universe, you would instantly know the state of the particle on the left side of the universe without having disturbed the left photon. Einstein, Podolsky, and Rosen rejected the idea that a measurement on one photon, here, could affect the state of the distant photon of the entangled paired. They argued that the consideration of the wave function for this system only was not sufficient and that quantum theory needed additional features to account for what they believed to be a necessary feature of the universe, local realism. As "the systems no longer interact", said the EPR team, "no real change can take place in the second system in consequence of anything that may be done to the first system."[125]

Bohr came back with a quick rebuttal having the same title: "Can quantum-mechanical description of physical reality be considered complete?"[126] Of this pair of papers, the physicist John Bell said, "I feel that Einstein's intellectual superiority over Bohr, in this instance, was enormous; a vast gulf between the man who saw clearly what was needed, and the obscurantist."[127]

Bell, moreover, recognized that the debate could be put to an experimental test,[128] that there was something that could be done in the laboratory to establish whether or not there might be additional so-called hidden variables that underlie the polarization state of a transmitted photon or any other quantum particle. The outcome of these experiments, raised to an art by Aspect, would give a posthumous victory either to Einstein or Bohr. The outcome would also, remarkably, determine whether the Law of Malus is sustainable. Thus, we have come full circle to the beginning of our journey, as any good story must reflect somehow on its beginning.

Bell derived inequalities, simple counting relationships of polarization states that should be obeyed by experiments having definite answers, that seem to be violated by quantum particles. It was recognized that these inequalities could be evaluated with simple-to-understand apparatuses of the kind employed by Aspect (Figure 12). While others had preceded Aspect in devising experiments to test Bell's inequalities, Aspect recognized that an important loophole needed to be closed.[129] In his apparatus, the state of the linear analyzers would be chosen only after the entangled particles had begun their flight. Thus, the photons would not be able to "conspire", to somehow communicate and coordinate their activities with superluminal communications. Aspect achieved this in-flight polarization scrambling by scattering the photons off ultrasonic wave gratings created in glass boxes of water excited by transducers.

The predictions of quantum mechanics, which turn out to be consistent with the Law of Malus, were shown by Aspect to hold firm, while Bell's inequalities were falsified. A deviation from the predictions of quantum theory would imply a more linear relation between the transmission of a photon and the angle between polarizers, than is the traditional cosine-squared relationship. Malus can rest in peace but in the absence of classical ideas of locality. *Here* is somehow connected to *there*, and in this story, *then* is somehow connected to *now*.

11. Conclusion

This discussion follows closely the presentation given by the author at the 16th International Conference on Chiroptical Spectroscopy. It is more of a personal history than a comprehensive history. Needless to say, important French figures have been omitted. Among these is Émile Verdet (1824-1866) who described unpolarized light and gave his name to the constants that describe Faraday rotation, Henri Poincaré (1854-1912) who developed a method for representing polarization states on the surface of a sphere, and Charles Fabry (1867-1945) who invented the Fabry-Pérot interferometer. We would be remiss if we failed to mention the great French liquid crystal pioneers and their spectacular applications of polarized light: Charles Mauguin (1878-1958), and Georges Friedel (1865-1933), already mentioned briefly within, as well as François Granjean (1882-1975). Of these omissions, *Que pouvez-vous faire*.

Acknowledgements

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GRAPHICAL ABSTRACT

Aimé Cotton's circular dichroism apparatus. From Recherches sur l'absorption et la dispersion de la lumiere par les milieux doues du pouvoir rotatoire. *J. Phys. Theor. Appl*. **1896**:5:237-244.

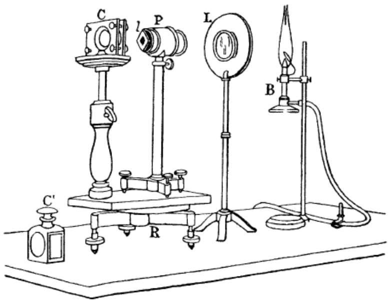

Fig. 1.

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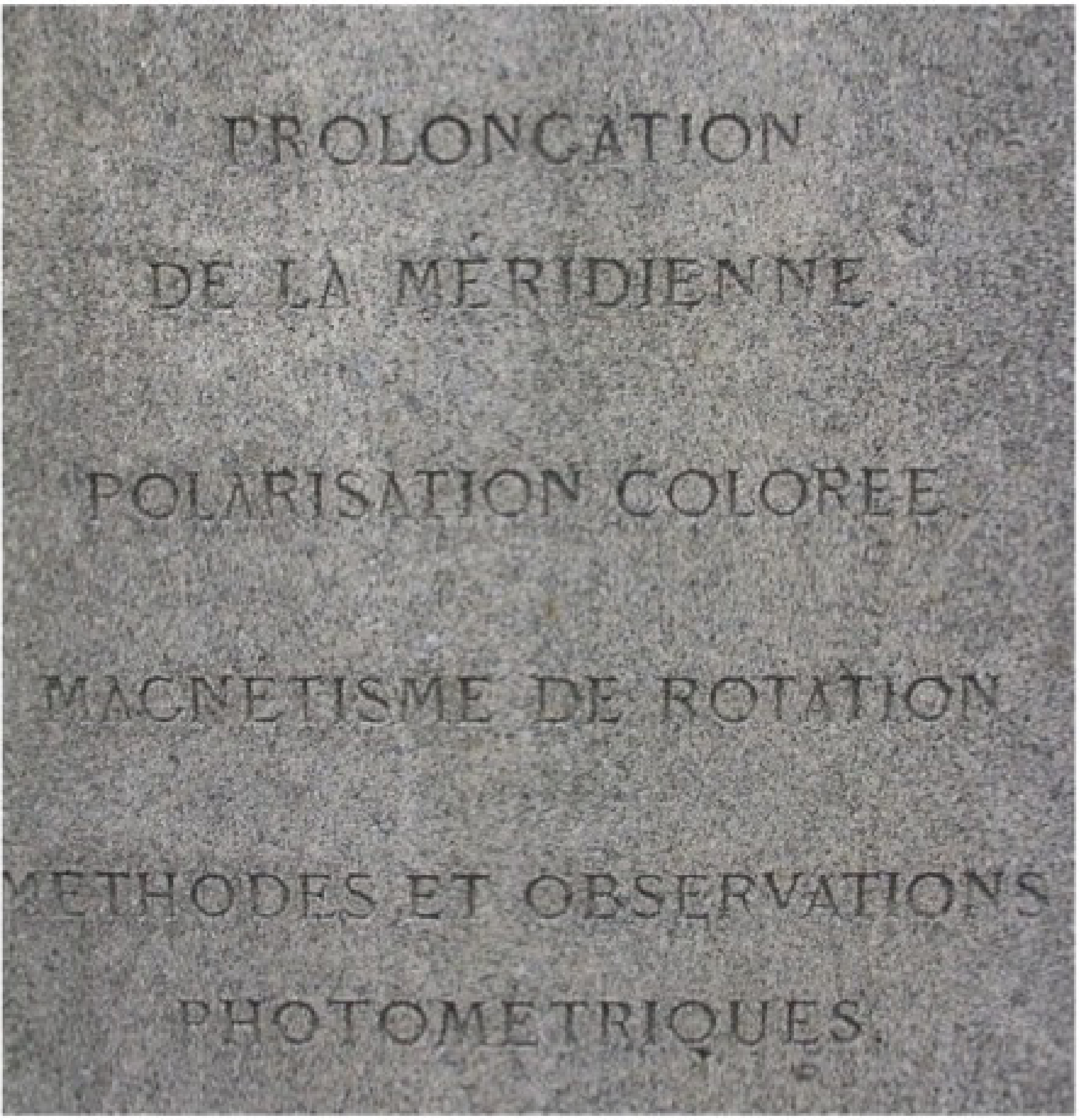

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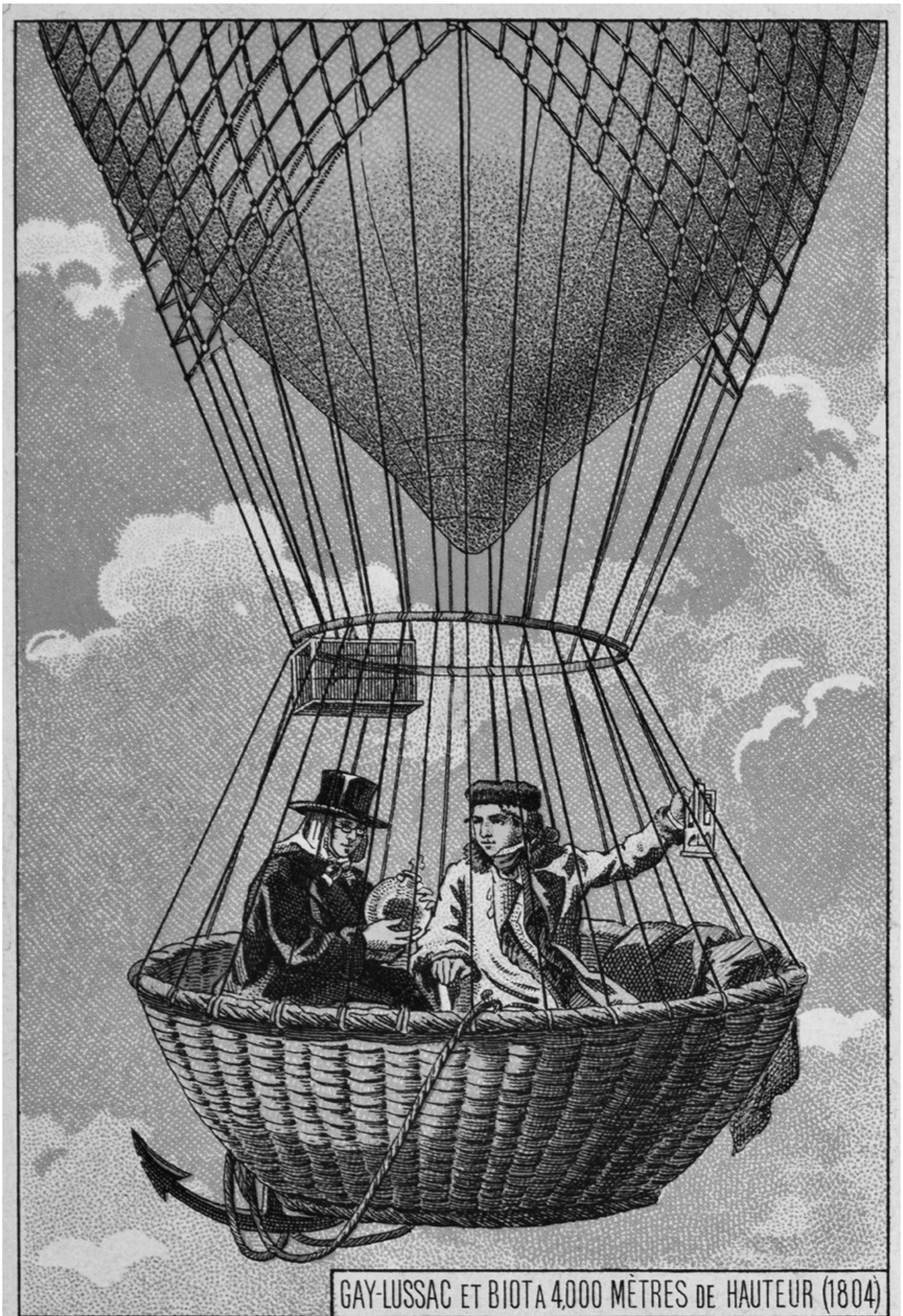

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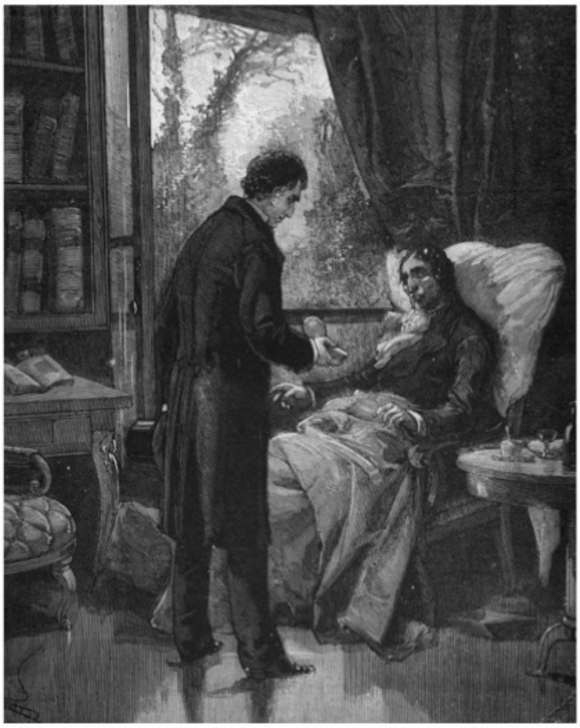

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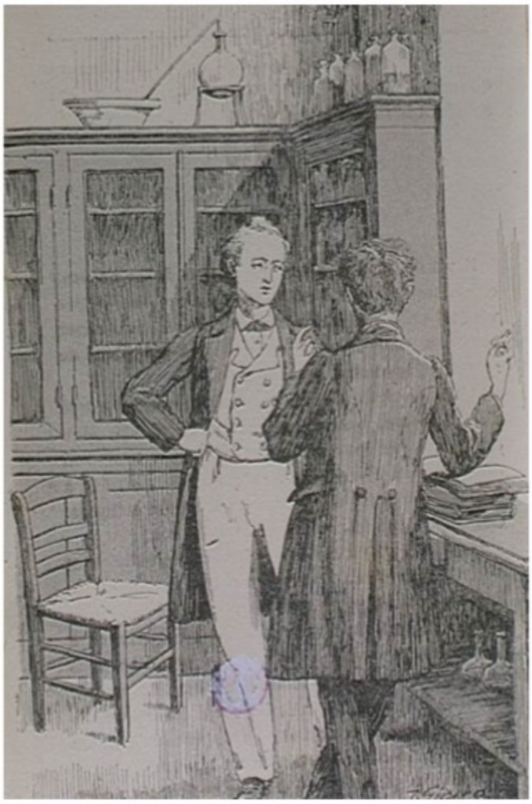

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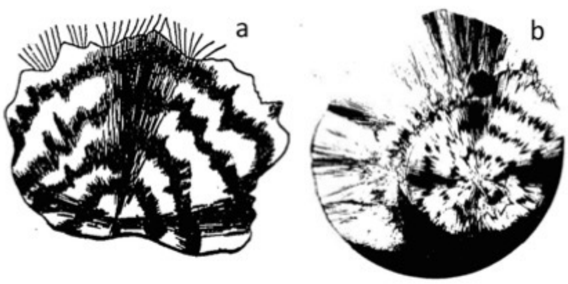

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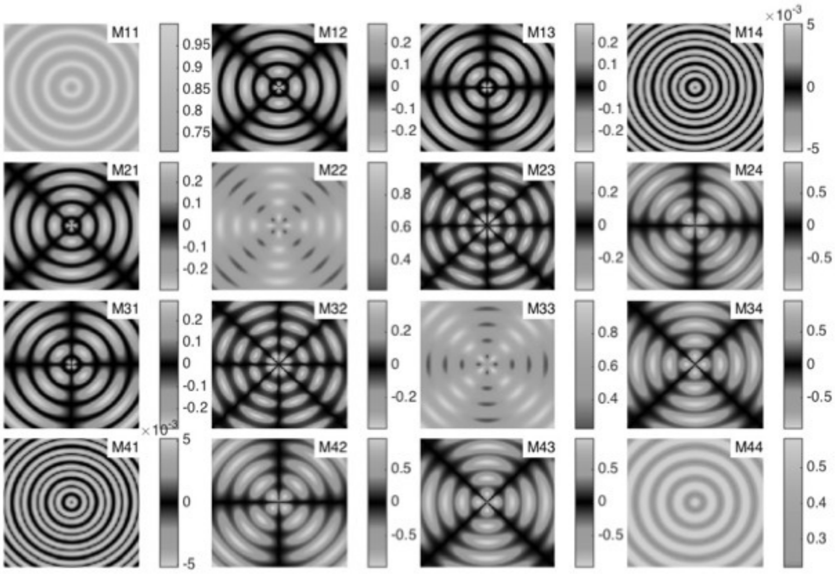

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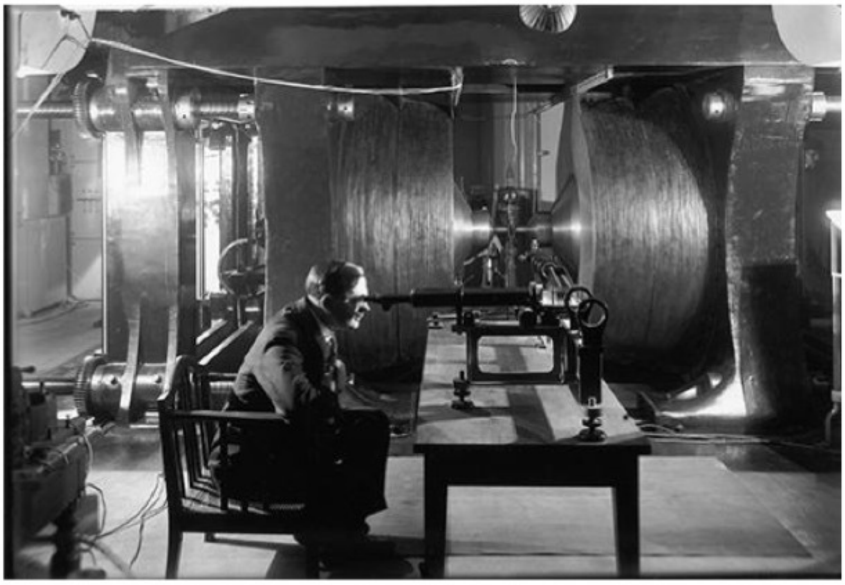

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SEMAINE DU 16 AU 22 AVRIL 1950

Pour remplacer Joliot l'atomique Zoé hésite entre six prétendants

« JAMAIS un véritable savant progressiste ne donnera une parcelle de sa science pour faire la guerre à l'U.R.S.S. »

Ces déclarations fracassantes de Joliot-Curie, au Congrès communiste de Gennevilliers, attirent de nouveau l'attention sur ce paradoxe. La France tolérera-t-elle plus longtemps que le maître de ses recherches atomiques se réclame d'un parti qui n'est plus français ?

Que M. Joliot-Curie choisisse entre son poste et le parti communiste, tel est le dilemme. Et puisqu'il s'obstine à ne pas vouloir le trancher lui-même, le gouvernement devra le trancher à sa place.

La succession de Joliot-Curie va donc s'ouvrir.

Des noms circulent, parmi lesquels le gouvernement aura à choisir un candidat qui ait à la fois des titres scientifiques indiscutables et une position politique moins marquée (ce n'est pas difficile !) que celle du camarade savant Joliot-Curie.

Bertrand GOLDSCHMIDT

Francis PERRIN

Jean THIBAUD

Louis DE BROGLIE

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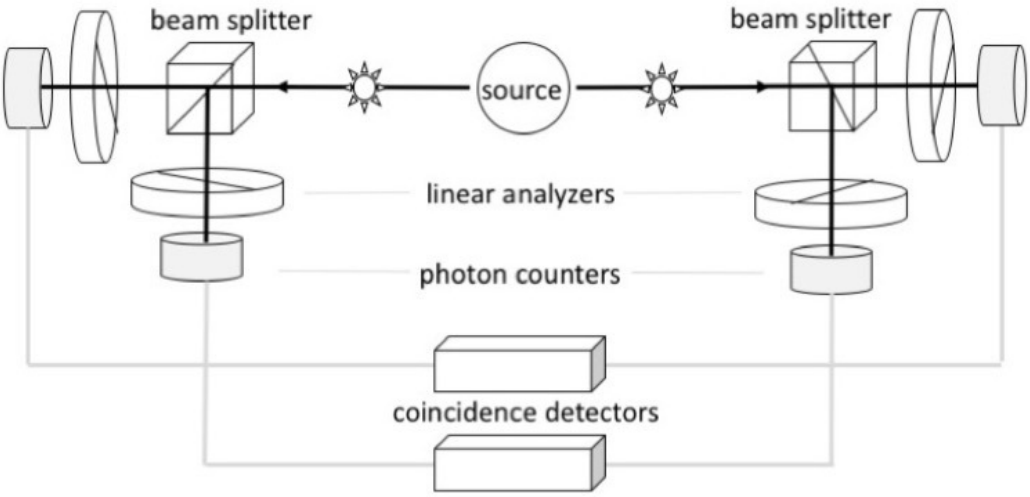

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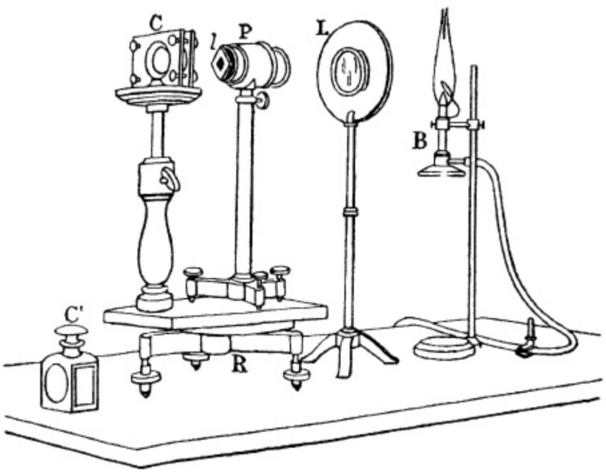

Fig. 1.

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