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Thermal decoherence in mesoscopic interference

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Abstract. A double-slit experiment is analysed when the interfering particle has an internal structure and one endeavours to obtain '*which path*' information by detecting the light that is spontaneously emitted. A compact expression is derived for the visibility of the interference pattern: coherence depends on both the spatial and temporal features of the emission process during the travel of the particle to the screen. A bound is set on the temperature of a fullerene molecule in a double-slit experiment in order that its quantum mechanical coherence be maintained.

1. Introduction

The present paper discusses the interference of *mesoscopic* systems. 'Mesoscopic' objects are neither microscopic nor macroscopic. They can be described by a wave function, yet are made up of a significant number of elementary constituents, such as atoms. Most importantly, they are characterized by a nontrivial internal structure that can have both quantal and classical features. A significant example, on which we shall focus our attention, is a molecule of fullerene, made up of 60 nuclei of carbon and 360 electrons, for a total of about 10^3 'elementary' constituents. Although fullerenes are fully quantum mechanical systems, they also have macroscopic-like features and emit thermal (blackbody) radiation [1–3]. Recently the quantum interference of fullerene molecules (C_{60} and C_{70}) has been observed in a series of pioneering experiments performed in Vienna [4, 5]. Our aim is to analyse the interference of fullerene from a theoretical viewpoint.

2. Double-slit interference

We start by looking at the simplest quantum mechanical experiment: consider a quantum system described by a wave packet impinging on a double slit. We assume that the wave packet travels along direction $+z$ and its transverse coherence length is larger than the separation of the slits, so that the two wave packets emerging from the slits are coherent with each other. The slits are parallel to y, have width a and are separated by a distance d, along direction x. The problem is essentially one-dimensional, the relevant coordinate being x.

Let the wave function emerging from one slit be $\langle x|\psi_{\rm slit}\rangle = \psi_{\rm slit}(x)$. The initial state at the double slit is

$$|\psi_0\rangle = \frac{1}{\sqrt{2}}(|\psi_+\rangle + |\psi_-\rangle), \qquad |\psi_\pm\rangle = \exp\left(\pm\frac{i}{\hbar}\frac{d}{2}p\right)|\psi_{\rm slit}\rangle, \tag{1}$$

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where p is the momentum operator and we assumed that the two wave packets do not overlap, $\langle\psi_+|\psi_-\rangle = 0$, hence $|\psi_0\rangle$ is normalized. Interference is observed at a screen perpendicular to z, placed at a distance $z = L$ from the plane of the slits. The intensity at the screen reads

$$I(x) = \langle\psi(t)|x\rangle\langle x|\psi(t)\rangle = |\psi(x,t)|^2, \qquad |\psi(t)\rangle = e^{-ip^2t/2m\hbar}|\psi_0\rangle, \tag{2}$$

where $t = L/v_z$ is the time of arrival of the wave packet at the screen. We shall focus on the experiment [5, 6] and take the slits to have width a and to be separated by a distance $d = 2a$. We set $d = 100\,\mathrm{nm}$, $L = 1.22\,\mathrm{m}$, $m = 1.197 \times 10^{-24}\,\mathrm{kg}$ and consider a beam with $\bar{v}_z = 128\,\mathrm{m/s}$, so that $\lambda_z = h/mv_z = 4.3\,\mathrm{pm}$ and $t = 9.47\,\mathrm{ms}$. In this case we are looking at the far-field interference pattern. Therefore, by taking the asymptotic limit $t \to \infty$ of $\psi(x,t)$ in (2), one gets

$$\begin{aligned}\psi(x,t) &= \int \frac{\mathrm{d}p}{(2\pi\hbar)^{\frac{1}{2}}}\tilde{\psi}_{\mathrm{slit}}(p)\sqrt{2}\cos\left(\frac{p\,d}{2\hbar}\right)e^{(-i/\hbar)(((p^2/2m)t)-px)} \\ &\sim \left(\frac{m}{it}\right)^{1/2}\tilde{\psi}_{\mathrm{slit}}\left(\frac{mx}{t}\right)e^{(i/\hbar)((mx^2)/(2t))}\sqrt{2}\cos\left(\frac{md}{2\hbar t}x\right),\end{aligned} \tag{3}$$

where $\tilde{\psi}_{\mathrm{slit}}(p) = \langle p|\psi_{\mathrm{slit}}\rangle$ is the Fourier transform of the wave packet emerging from one slit. Using equation (3), the intensity pattern (2) reads

$$I(x) = I_{\mathrm{slit}}(x)\left[1 + \cos\left(2\pi\frac{x}{X}\right)\right], \tag{4}$$

$$I_{\mathrm{slit}}(x) = \frac{m}{t}\left|\tilde{\psi}_{\mathrm{slit}}\left(\frac{mx}{t}\right)\right|^2, \qquad X = \frac{ht}{md} = \frac{\lambda_z L}{d} = 52.46\,\mu\mathrm{m}. \tag{5}$$

Notice that the envelope function in (5) is correctly normalized, namely $\int \mathrm{d}x\, I_{\mathrm{slit}}(x) = \int \mathrm{d}p\, |\tilde{\psi}_{\mathrm{slit}}(p)|^2 = 1$.

3. Two-level molecule

So far the interfering system has been considered as a structureless particle. However, we aim at describing a more complicated physical picture, that can arise when the interfering system is endowed with a richer internal physical structure. In order to obtain '*which path*' information, one might shine laser light on the molecule after it has gone through the slits, as with the Heisenberg–Bohm microscope [6, 7] and look at the scattered photon. However, the situation here would be different, because the internal state is excited by the laser and the molecule spontaneously emits a photon with a given lifetime during its travel to the screen.

The minimal requirement on λ_0 to maintain quantum coherence and preserve the interference pattern is the Heisenberg condition [6]

$$\lambda_0 \gtrsim 2d. \tag{6}$$

However, we shall see that, at variance with the structureless case, this is not the only criterion.

Consider the Hamiltonian

$$H = H_0 + V, \qquad H_0 = \frac{\boldsymbol{p}^2}{2m} + \hbar\omega_0 |e\rangle\langle e| + \sum_i \hbar\omega_i a_i^\dagger a_i,$$
$$V = \sum_i \left(\Phi_i e^{i\boldsymbol{k}_i\cdot\boldsymbol{x}} |e\rangle\langle g| a_i + \text{h.c.}\right), \tag{7}$$

describing a two-level system of mass m, position $\boldsymbol{x}$ and momentum $\boldsymbol{p}$, coupled to the electromagnetic field, $[a_i, a_j^\dagger] = \delta_{ij}$. The ground state $|g\rangle$ has energy 0, while the excited state $|e\rangle$ has energy $\hbar\omega_0$. The state of the total system will be written $|\Psi_{\text{tot}}\rangle = |\psi, \alpha, n_i\rangle \equiv |\psi\rangle \otimes |\alpha\rangle \otimes |n_i\rangle$, where $|\psi\rangle$ denotes the spatial part of the wave function of the molecule, $\alpha = e, g$ and n_i is the number of photons emitted in the i-mode.

Due to laser excitation, the molecule emerges from the two slits in an excited state $|\psi_0, e, 0\rangle$, where $|\psi_0\rangle$ is given in equation (1). The evolution of the spontaneous emission process is readily computed in the Weisskopf–Wigner approximation [10] and yields [11]

$$|\Psi_{\text{tot}}(t)\rangle = e^{-i\omega_0 t} e^{-\gamma t/2} |\psi(t), e, 0\rangle + \sum_i e^{-i\omega_i t} \beta_i(t) |e^{-i\boldsymbol{k}_i\cdot\boldsymbol{x}}\psi(t), g, 1_i\rangle, \tag{8}$$

$$\beta_i(t) = \frac{\Phi_i^*}{\hbar} \frac{1 - e^{i(\omega_i - \omega_0)t - \gamma t/2}}{(\omega_i - \omega_0) + i\gamma/2}, \tag{9}$$

where $|\psi(t)\rangle$ is given by (9) and $\gamma = 2\pi \sum_i \delta(\omega_i - \omega_0)|\Phi_i|^2/\hbar^2$ is the decay rate, as given by the Fermi golden rule. We see that in (8) the internal degrees of freedom of the molecule get entangled with the photon field, so that the states in (8) are all orthogonal to each other. Notice that $\sum_i |\beta_i(t)|^2 = 1 - e^{-\gamma t}$, hence $|\Psi_{\text{tot}}(t)\rangle$ is normalized at every time. By assuming that the spontaneous emission process is completely isotropic, the intensity at the screen reads

$$\begin{aligned} I'(x) &= \langle \Psi_{\text{tot}}(t)|x\rangle\langle x|\Psi_{\text{tot}}(t)\rangle \\ &= \exp(-\gamma t) I(x) + (1 - e^{-\gamma t}) \int \frac{\mathrm{d}\Omega_{\boldsymbol{n}}}{4\pi}\, I_{-\boldsymbol{n}\omega_0/c}(x), \end{aligned} \tag{10}$$

where $\mathrm{d}\Omega_{\boldsymbol{n}}$ is the differential solid angle, $\boldsymbol{n}$ a unit vector. The interference pattern (10) is made up of two terms: the first one is associated with those molecules that have not emitted any photons, the second one with those molecules that have emitted a photon and recoiled accordingly. The quantity $I_{-\boldsymbol{n}\omega_0/c}(x)$ represents the partial interference pattern of those molecules that have emitted a photon of momentum $\hbar\boldsymbol{k} = \hbar\boldsymbol{n}\omega_0/c$ and reads

$$I_{-\boldsymbol{n}\omega_0/c}(x) = I(x - v_x t), \tag{11}$$

where $v_x = \hbar k_x/m$ is the x-component of the recoil velocity. By (11) and (4), the average over the direction of the emitted photon yields

$$\begin{aligned} \int \frac{\mathrm{d}\Omega_{\boldsymbol{n}}}{4\pi}\, I_{-\boldsymbol{n}\omega_0/c}(x) &= I_{\text{slit}}(x) \int_{-1}^{1} \frac{\mathrm{d}\xi}{2} \left[1 + \cos\left(\frac{2\pi}{X} x - \frac{\omega_0 d}{c}\xi\right)\right] \\ &= I_{\text{slit}}(x) \left[1 + \operatorname{sinc}\left(\frac{\omega_0 d}{c}\right) \cos\left(\frac{2\pi}{X} x\right)\right], \end{aligned} \tag{12}$$

where $\operatorname{sinc}(x) \equiv \sin x/x$ and we used the equality $2\pi\hbar\omega_0 t/mcX = \omega_0 d/c$. It is evident from this expression that when $\omega_0 d/c = \pi$, i.e. $\lambda_0 = 2\pi c/\omega_0 = 2d$ the

cosine is averaged over the whole interval 2π and interference term is completely washed out, in agreement with the Heisenberg condition (6). For larger values of λ_0 there is still some interference. By plugging (12) into (10) we finally obtain

$$I'(x) = I_{\mathrm{slit}}(x)\left[1 + \mathcal{V}\left(\gamma t, \frac{d}{\lambda_0}\right)\cos\left(\frac{2\pi}{X}x\right)\right], \tag{13}$$

$$\mathcal{V}\left(\gamma t, \frac{d}{\lambda_0}\right) = e^{-\gamma t} + (1 - e^{-\gamma t})\,\mathrm{sinc}\left(\frac{2\pi d}{\lambda_0}\right). \tag{14}$$

The interpretation of the visibility $\mathcal{V}$ derives from (10): the first term in the r.h.s. of (14) is associated with those molecules that have not emitted any photon (and reach the screen in an excited state), while the second term is associated with those molecules that have emitted a photon before they hit the screen.

Let $\lambda_0 < 2d$, so that the coherence condition (6) is not satisfied. Nevertheless, we see from (14) that coherence is still largely preserved if $\gamma t \lesssim 1$, because even though the photon wavelength is small enough to yield information about the path of the interfering particle, such path information is not accessible: it is, so to say, stored in the internal structure of the molecule. Such information would be available to an external observer only if the photon were emitted. Mathematically,

$$\mathcal{V}\left(\gamma t, \frac{d}{\lambda_0} \gtrsim \frac{1}{2}\right) \simeq \mathcal{V}(\gamma t, \infty) = \exp(-\gamma t), \tag{15}$$

which tends to vanish if the decay is rapid ($\gamma t \gtrsim 1$) and to unity if the decay is slow ($\gamma t \lesssim 1$). In conclusion, the interference pattern is blurred out ($\mathcal{V} \simeq 0$), only if the photon emission process yields *both* a good resolution, $\lambda_0 \lesssim 2d$, *and* a quick response, $\gamma t \gtrsim 1$.

4. Fullerene molecule

A molecule of fullerene is a mesoscopic system, that can absorb several visible photons at once and undergo quite involved processes in its internal structure, involving lifetimes, emission of blackbody radiation [2, 3] and complex ionization processes [1, 8, 9]. In this section we consider the fullerene as a small black body, that starts its evolution, immediately after the slits, in a highly excited state characterized by a well-defined temperature. On its way to the screen, it emits $N = n \pm \triangle n$ photons of different energies and in random directions. As a consequence the momentum of the molecule will recoil by the quantity $\triangle p \simeq \hbar\langle k\rangle\sqrt{n}$, where $\hbar\langle k\rangle$ is the average momentum of the emitted photons and n their mean number. On the other hand the molecule loses a total energy $\triangle E \simeq n\hbar\langle k\rangle/c$ between the slits and the screen. Hence, its momentum will be changed by the quantity

$$\triangle p \simeq \frac{\triangle E}{c\sqrt{n}}. \tag{16}$$

Therefore the interference pattern will be only slightly affected by the emission of a large number of low-energy photons. This is an interesting qualitative conclusion.

A more quantitative relation can be obtained by treating the fullerene molecule as a 'macroscopic' system that emits thermal radiation at temperature T. The

energy and number of photons emitted by the surface A of the fullerene molecule during its time of flight t is, respectively, [12]

$$\triangle E = \alpha J_0 A t, \qquad n = \alpha \Phi_0 A t, \qquad J_0 = \frac{\pi^2}{60}\frac{k_B^4}{c^2\hbar^3}T^4, \qquad \Phi_0 = \frac{\zeta(3)}{2\pi^2}\frac{k_B^3}{c^2\hbar^3}T^3, \quad (17)$$

where k_B is the Boltzmann constant, $\zeta(3) \simeq 1.202$ the Riemann function and $\alpha \simeq 4.5 \times 10^{-5}$ an emissivity coefficient, due to the curvature of the emitting fullerene surface [1, 3, 8]. Therefore by making use of (17), equation (16) reads

$$\triangle p \simeq \frac{\alpha J_0 A t}{c\sqrt{\alpha \Phi_0 A t}} = \sqrt{\alpha}\kappa (At)^{1/2} T^{5/2}, \qquad \kappa = \sqrt{\frac{2}{\zeta(3)}}\frac{\pi^3}{60}\frac{k_B^{5/2}}{c^2\hbar^{3/2}}. \quad (18)$$

In order to observe interference, the transferred momentum $\triangle p$ must satisfy the coherence condition (6)

$$\triangle p = \frac{h}{\lambda_0} \lesssim \frac{h}{2d}, \quad (19)$$

which translates into the following bound for the internal temperature T

$$T \lesssim T_{\rm dec} \equiv \xi \alpha^{-1/5} \frac{1}{(Atd^2)^{1/5}}, \quad (20)$$

$$\xi = \left(\frac{1800\ \zeta(3)}{\pi^4}\right)^{1/5} \frac{\hbar\ c^{4/5}}{k_B} = 8.59 \times 10^{-5} \mathrm{Ks}^{1/5}\ \mathrm{m}^{4/5}. \quad (21)$$

Equation (20) is a *coherence condition*. The quantity $T_{\rm dec}$ is the internal (blackbody) temperature of a fullerene molecule at which decoherence effects should become apparent in a double slit experiment. For the numerical values of the Vienna experiment, by taking $A = 4\pi r^2 = 1.539 \times 10^{-18}$m ($r \simeq 3.5$ Å is the fullerene radius) and $t = 9.47$ms [5, 9] we get $T_{\rm dec} \simeq 3700$ K. Notice that above $T \simeq 3000$ K fullerene molecules begin to fragmentate (ionization is likely to occur at even lower temperatures). Therefore, the temperature of the fullerene molecule will have only a small influence on the visibility of the interference pattern, at least for the Vienna experimental configuration. However, if the experiment is modified by letting the fullerene pass through an interferometer of the Mach–Zender type in order to increase the beam separation d, then intrinsic decoherence effects should come to light. The behaviour of $T_{\rm dec}$ versus d (slit separation) is shown in figure 1 for a time of flight of 9.53 ms [5]. Decoherence effects should be visible at about 2000 K for a beam separation of the order of half a micron.

5. Conclusions

We have studied the double-slit interference pattern and the coherence properties of a molecule with an increasingly complicated internal structure.

If the molecule is 'elementary', i.e. structureless, the usual description in terms of the Heisenberg–Bohm microscope applies and interference is lost when a photon of suitable wavelength is scattered off the molecule after the latter has gone through a double slit.

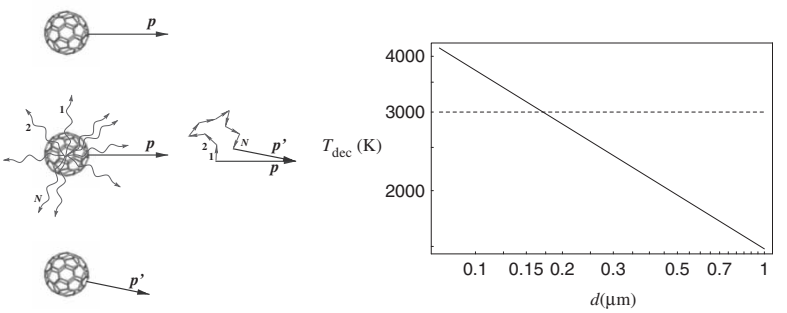

Figure 1. Random walk in momentum space; N (thermal) photons are emitted and the momentum of the molecule changes accordingly. T_{dec} versus d (slit separation) for a fullerene molecule; we set the time of flight $t = 9.53\,\mathrm{ms}$; the horizontal line is the temperature at which fragmentation becomes significant.

If, on the other hand, the molecule has an internal structure (for example a two-level system), one still needs a photon of suitable wavelength to destroy interference, but in addition the photon re-emission process must be rapid. If, for instance, the photon is re-emitted only after the molecule has reached the screen, no '*which path*' information is available and interference (coherence) is preserved.

Finally, if the molecule can absorb and re-emit a large number of photons and is complicated enough to have an internal *temperature*, one can properly talk of 'mesoscopic interference'. In such a situation, one can obtain sensible results by combining thermodynamical considerations with a pure quantum mechanical analysis. The mesoscopic system will slowly 'explore' its environment by emitting photons in the course of its evolution and its branch waves will slowly 'decohere' (namely, they get entangled with increasingly orthogonal states of the electromagnetic field, that plays the role of environment). In this sense, coherence—viewed as ability to interfere—simply means isolation from the environment.

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