## Interference of mesoscopic particles: quantum-classical transition

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We analyze the double slit interference of a mesoscopic particle. We calculate the visibility of the interference pattern, introduce a characteristic temperature that defines the onset to decoherence and scrutinize the conditions that must be satisfied for an interference experiment to be possible.

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<u>Introduction</u> - Interference is one of the most characteristic traits of quantum systems. As Dirac clarified [1], this phenomenon is rooted in the superposition principle, according to which different states of a single quantum mechanical particle interfere with each other. The simplest case is that of two states: double slit interference has been observed with photons, electrons, neutrons, atoms and small molecules [2], and recently even with large molecular clusters [3]. Our comprehension of the quantum mechanical world has been shaped, to a large extent, by the ideas that motivated these experiments.

Quantum particles interfere, but classical particles do not, and it is not easy to understand where the borderline has to be placed. The size of the interfering system plays an important role, but it is certainly not the only relevant variable: for example, double slit interference has been observed with molecules, but not with protons. In this Letter we will analyze the interference of mesoscopic systems, endowed with an internal structure which leads to entanglement with their environment (e.g., via photon emission). An example is a fullerene molecule flying between a diffraction grating and a detector. Some classical features are apparent for such mesoscopic systems, vet their ability to interfere is preserved, at least to some extent, and can be viewed as a quantum signature. The main objective of this Letter will be to understand under which conditions a "large" system interferes and which ones of its dynamical variables can interfere.

Double slit interference in a Poissonian environment -Let us consider a mesoscopic quantum system (molecule), whose center of mass is described by a (double) wave packet  $|\psi_0\rangle$ , emerging from a double slit. The wave packet travels along direction +z; the slits are parallel to y and are separated by a distance d, along direction x $(\hbar = 1)$ :

$$|\psi_{0}\rangle = \frac{1}{\sqrt{2}}(|\psi_{+}\rangle + |\psi_{-}\rangle),$$
  
$$|\psi_{\pm}\rangle = \exp\left(\pm i\frac{d}{2}p_{x}\right)|\psi_{\text{slit}}\rangle, \qquad (1)$$

where  $p_x$  is the x component of the momentum operator and  $|\psi_{\text{slit}}\rangle$  the state emerging from one slit. We assume that  $\langle \psi_{-} | \psi_{+} \rangle = 0$ , so that  $| \psi_{0} \rangle$  is normalized. During its travel to the screen, the molecule emits photons and recoils accordingly. The internal state of the molecule together with the photon field plays therefore the role of environment: such an environment disturbs the motion of the center of mass, via scattering processes (typically photon emissions, yielding momentum kicks). We shall assume that the internal temperature of the mesoscopic system is much higher than the temperature of the photon field.

Let the molecule undergo momentum kicks ( $\Delta p_i, i \in \mathbb{Z}$ ) due to photon emissions. The Hamiltonian describing the evolution of the x component of the center of mass in the presence of random kicks  $\Delta p_k$  at times  $t_k$  reads (henceforth, for simplicity,  $p_x = p$ )

$$H_{\xi}(t) = \frac{p^2}{2m} - \xi(t)x,$$
 (2)

$$\xi(t) = \sum_{k} \delta(t - t_k) \Delta p_k, \qquad (3)$$

where  $\{t_k\}_{k\in\mathbb{Z}}$  is a shot noise with density  $\Lambda$  and the momentum jumps  $\Delta p_k$  are independent identically distributed random variables with probability density  $W(\Delta p_k)$ . The process  $\xi(t)$  is the time derivative of a compound Poisson process [4]. Both  $\Lambda$  and W are functions of the state of the environment (for example its temperature of T).

The time evolution of a wave packet which emerges at time  $t_0 = 0$  from the slits reads

$$|\psi(t)\rangle = U_{\xi}(t)|\psi_0\rangle,\tag{4}$$

 $U_{\xi}(t)$  being the unitary evolution engendered by the Hamiltonian (3)

$$U_{\xi}(t) = \mathcal{T} \exp\left(-i \int_{0}^{t} ds \ H_{\xi}(s)\right)$$
$$= e^{-i(t-t_{n})p^{2}/2m} \mathcal{T} \prod_{k=1}^{n} e^{ix \bigtriangleup p_{k}} e^{-i\bigtriangleup t_{k-1}p^{2}/2m}$$
$$= e^{ix \bigtriangleup p^{(n)}} \prod_{k=0}^{n} e^{-i(p-\bigtriangleup p^{(k)})^{2} \bigtriangleup t_{k}/2m}, \tag{5}$$

where  $\Delta t_k = t_{k+1} - t_k$ ,  $\Delta p^{(k)} = \sum_{j=1}^k \Delta p_j$  is the total effect of k momentum jumps ( $\Delta p^{(0)} = 0$ ), the total number of collisions n is a Poisson random variable with mean  $\Lambda t$ , and  $\mathcal{T}$  is the time-ordering operator, forcing earlier times (lower k) at the right. In the third equality we used the commutation relation  $e^{-ip^2 \Delta t/2m} e^{ix\Delta p} = e^{ix\Delta p} e^{-i(p-\Delta p)^2 \Delta t/2m}$ , in order to move all kick operators to the far left side.

From (4) and (5) one gets

$$\psi(x,t) = \langle x|\psi(t)\rangle$$
  
=  $e^{ix \triangle p^{(n)}} \langle x|e^{-i\sum(p-\triangle p^{(k)})^2 \triangle t_k/2m}|\psi_0\rangle$   
=  $e^{ix \triangle p^{(n)}} \int \frac{dp}{\sqrt{2\pi}} e^{-i\phi(p)} \tilde{\psi}_0(p),$  (6)

where  $\tilde{\psi}_0(p) = \langle p | \psi_0 \rangle$  and

$$\phi(p) = \sum_{k=0}^{n} \frac{(p - \triangle p^{(k)})^2 \triangle t_k}{2m} - xp$$
(7)

is a quadratic polynomial in p with quadratic term  $p^2 \sum \Delta t_k/2m = p^2 t/2m$ . It can be rewritten as

$$\phi(p) = \phi(\bar{p}) + \frac{t}{2m}(p - \bar{p})^2,$$
(8)

 $\bar{p}$  being the value of the momentum at the extremal  $\phi'(\bar{p}) = 0$ , that is

$$\bar{p}(x,t) = \frac{mx}{t} + \sum_{k=0}^{n} \frac{\triangle p^{(k)} \triangle t_k}{t} = \frac{mx}{t} + \sum_{k=1}^{n} \zeta_k \triangle p_k, \quad (9)$$

where  $\zeta_k = 1 - t_k/t$  characterize the emissions between the grating and screen. A peculiarity of this analysis is the presence of the *same* Poisson process on *both* branch waves: for an external environment, one should have considered *two independent* Poisson processes, one for each branch wave.

Equation (6) represents the convolution of the initial momentum wave packet with a Gaussian

$$\psi(x,t) = e^{i\left[x \triangle p^{(n)} - \phi(\bar{p})\right]} \int \frac{dp}{\sqrt{2\pi}} e^{-i\frac{t}{2m}(p-\bar{p})^2} \tilde{\psi}_0(p), \ (10)$$

whose spread  $(m/t)^{1/2}$  becomes narrower as time t increases. For  $t \to \infty$  (10) reads

$$\psi(x,t) \sim e^{i\left[x \bigtriangleup p^{(n)} - \phi(\bar{p})\right]} \left(\frac{m}{it}\right)^{\frac{1}{2}} \tilde{\psi}_0(\bar{p}). \tag{11}$$

This approximation is valid for  $t \gg m |\tilde{\psi}_0''(\bar{p})/\tilde{\psi}_0(\bar{p})|$  and implies that  $\tilde{\psi}_0(p)$  can be represented by the constant value  $\tilde{\psi}_0(\bar{p})$  on the screen. In the following we will always suppose that such condition holds (far field interference pattern). The interference pattern reads

$$I(x,t) = \left\langle |\psi(x,t)|^2 \right\rangle \sim \left(\frac{m}{t}\right) \left\langle |\tilde{\psi}_0(\bar{p})|^2 \right\rangle$$
(12)

where  $\langle \cdots \rangle$  denotes the average over the process  $\xi(t)$ . With the initial state (1),  $\tilde{\psi}_0(p) = \sqrt{2}\tilde{\psi}_{\rm slit}(p)\cos(pd/2)$ , with  $\tilde{\psi}_{\rm slit}(p) \equiv \langle p|\psi_{\rm slit}\rangle$ , and the far-field condition is satisfied for  $t \gg md^2$ . Under this condition, the intensity at the screen reads

$$I(x,t) = \left(\frac{m}{t}\right) \left| \tilde{\psi}_{\text{slit}} \left(\frac{mx}{t}\right) \right|^2 \left[ 1 + \left\langle \cos\left(\bar{p}d\right) \right\rangle \right], \quad (13)$$

where we approximated  $|\tilde{\psi}_{\rm slit}(\bar{p})|^2 \simeq |\tilde{\psi}_{\rm slit}(mx/t)|^2$ , for weak enough kicks. The corresponding visibility  $\mathcal{V}$  is

$$I(x,t) = I_0(x,t) \left[ 1 + \mathcal{V} \cos\left(\frac{md}{\hbar t}x + \phi\right) \right],$$
  

$$\mathcal{V} = |F|, \quad \phi = \arg F,$$
  

$$F = \left\langle \exp\left(id\sum_{k=1}^n \zeta_k \triangle p_k\right) \right\rangle = \mathcal{V}e^{i\phi}, \quad (14)$$

where  $I_0(x,t) = (m/t)|\tilde{\psi}_{\rm slit}(mx/t)|^2$ . In order to calculate the visibility, the features of the average  $\langle \cdots \rangle$  must be expressed in terms of the distribution of the momentum jumps  $W(\Delta p_k)$  and the Poisson times  $\{t_k\}$ .

By performing first the average over  $W(\triangle p_k)$  we get

$$F = \left\langle \prod_{k=1}^{n} g(t_k) \right\rangle_{\Lambda}, \qquad (15)$$

where  $g(t_k) = f\left(d\frac{t-t_k}{t}\right)$  and

$$f(x) = \langle \exp(ix \Delta p) \rangle_{\Delta p} = \int du \ W(u) \exp(ixu), \quad (16)$$

 $\langle \cdots \rangle_{\Lambda}$  denoting the average over the shot noise with density  $\Lambda$ . This is easily computed [5]

$$F = 1 + \sum_{n=1}^{\infty} \frac{1}{n!} \left[ \Lambda \int_{0}^{t} (g(\tau) - 1) d\tau \right]^{n}$$
  
=  $\exp\left( -\Lambda t \int_{0}^{1} [1 - f(sd)] ds \right),$  (17)

and yields the visibility (Re denotes the real part)

$$\mathcal{V} = \exp(-\Lambda\zeta t), \quad \zeta = \int_0^1 \left[1 - \operatorname{Re} f(sd)\right] ds.$$
 (18)

By using the definition (16), the "geometrical" factor reads  $\zeta = \langle 1 - \operatorname{sinc}(d \Delta p) \rangle_{\Delta p}$ , where  $\operatorname{sinc} x = \sin x/x$ , and the visibility (18) can be given the useful expression

$$\mathcal{V} = \exp\left(-\Lambda t \left\langle 1 - \operatorname{sinc}(d \triangle p) \right\rangle_{\triangle p}\right).$$
(19)

Notice that  $\mathcal{V} \leq 1$ , because  $\operatorname{sinc} x \leq 1$ . Moreover, if the jumps are symmetrically distributed, i.e.  $W(\Delta p) = W(-\Delta p)$ , then f(x) is a real function and one can omit the real part in (18), so that  $\mathcal{V} = F$ .

<u>Thermodynamics</u> - In order to calculate the visibility from Eq. (19) we need to evaluate the kick rate  $\Lambda$  and the probability density of the momentum jumps  $W(\Delta p)$ . Planck's blackbody formula is not valid for small atomic clusters and needs to be generalized on two counts. One is the finiteness of the number of modes (freedoms), the other is the reduced stimulated emission. The former influences the high-energy part of the spectrum, the latter the low-energy part. In addition, there are finite-size effects that need to be taken into account. These are usually dealt with heuristically.

If the interfering cluster can be considered (almost) isolated during its travel to the screen, its temperature is in general not in equilibrium with that of the background radiation field. When photon absorption from the background radiation can be neglected (which is the case in which we are interested), the photon emission rate reads [6]

$$R_T(\omega) = \frac{\omega^2 \sigma_{\rm abs}(\omega)}{\pi^2 c^2} \exp\left[-\frac{\hbar\omega}{k_B T} - \frac{k_B}{2C_V} \left(\frac{\hbar\omega}{k_B T}\right)^2\right],\tag{20}$$

where  $\omega$  is the photon frequency,  $\sigma_{\rm abs}$  and  $C_V = Nk_B$ are the absorption cross section and heat capacity of the small particle, respectively, *T* its temperature and *N* the number of vibrational modes (for example,  $N \simeq 170$  for  $C_{60}$ ,  $N \simeq 200$  for  $C_{70}$ ).

The total photon rate reads

$$\Lambda(T) = \int_0^\infty d\omega \ R_T(\omega), \qquad (21)$$

where we assume that the temperature of the molecule does not change appreciably due to photon emission during the flight. We will check the validity of this assumption later. In order to compute these quantities, we have to determine the  $\omega$  dependence of the absorption cross section. We shall heuristically assume the form

$$\sigma_{\rm abs}(\omega) = a_\ell \omega^\ell, \tag{22}$$

where  $\ell$  is a positive integer and  $a_{\ell}$  a real number, and look for the best fit. For instance, in the case of the fullerenes C<sub>60</sub> and C<sub>70</sub>, a comparison with experiment [7] yields accurate fits for  $a_4 = 7.04 \times 10^{-66} \text{nm}^2 s^4$  and  $a_4 = 7.79 \times 10^{-66} \text{nm}^2 s^4$ , respectively [8]. By plugging Eq. (22) into (20) one gets the series

$$R_T(\omega) = \frac{a_\ell \omega^{\ell+2}}{\pi^2 c^2} e^{-\frac{\hbar\omega}{k_B T}} \sum_{m=0}^{\infty} \frac{(-1)^m}{(2N)^m m!} \left(\frac{\hbar\omega}{k_B T}\right)^{2m}$$
(23)

and integrating term by term in Eq. (21) one gets the asymptotic expansion for large  ${\cal N}$ 

$$\Lambda(T) \sim \frac{a_{\ell}}{\pi^2 c^2} \left(\frac{k_B T}{\hbar}\right)^{\ell+3} \sum_{m=0}^{\infty} \frac{(-1)^m (2m+\ell+2)!}{(2N)^m m!}.$$
(24)

Typical emission rates at T = 2500 for a time of flight  $t \simeq 2$  ms yields 4-5 emitted infrared photons during the

flight in the interferometer. In such case, the temperature of the molecule decreases by just a few percent, which does not affect the emission rate, and corroborates our initial assumption [after Eq. (21)].

The momentum kick on the molecule after the emission of a photon of frequency  $\omega$  has magnitude  $|\mathbf{p}| = \hbar \omega / c$ . By assuming that the emission process is isotropic, the probability density that the molecule undergoes a momentum jump  $\mathbf{p}$  reads

$$W^{(3D)}(\boldsymbol{p}) = \frac{1}{4\pi \boldsymbol{p}^2} \frac{c}{\hbar \Lambda(T)} R_T \left(\frac{c|\boldsymbol{p}|}{\hbar}\right), \qquad (25)$$

from which the one-dimensional probability density can be evaluated

$$W(\Delta p) = \int d^3 p \ W^{(3D)}(\mathbf{p}) \delta(p_x - \Delta p)$$
$$= \frac{c}{2\hbar\Lambda(T)} \int_{c|\Delta p|/\hbar}^{\infty} \frac{d\omega}{\omega} R_T(\omega).$$
(26)

By plugging (26) into (16) we get

$$f(x) = \frac{1}{\Lambda(T)} \int_0^\infty d\omega R_T(\omega) \operatorname{sinc}\left(\frac{\omega x}{c}\right), \qquad (27)$$

and from (18)

$$\mathcal{V}(T,d,t) = \exp\left(-[\Lambda(T) - G(T,d)]t\right)$$
(28)

where

$$G(T,d) = \int_0^1 ds \int_0^\infty d\omega R_\omega(T) \operatorname{sinc}\left(\frac{\omega d}{c}s\right).$$
(29)

Note that  $G(T,d) \sim \Lambda(T)$ , for  $T \to 0$ , so that for low temperatures  $\mathcal{V} \to 1$ . In the high temperature case, on the contrary,  $\mathcal{V} \to 0$ , as expected for a classical particle.

By plugging the series (23) into (29) and integrating term by term one gets

$$G(T,d) \sim \frac{a_{\ell}}{\pi^2 c d} \left(\frac{k_B T}{\hbar}\right)^{\ell+2} \sum_{m=0}^{\infty} \left\{ \frac{(-1)^m (2m+\ell+1)!}{(2N)^m m!} \times \int_0^{\frac{dk_B T}{\hbar c}} dx \frac{\sin\left[(2m+\ell+2)\arctan x\right]}{x(1+x^2)^{(2m+\ell+2)/2}} \right\}.$$
(30)

Visibility and quantum-classical transition - By inserting the expansions (24) and (30) (with  $\ell = 4, a_4 =$  $7.79 \times 10^{-66}$  [8]) into the visibility (28) one gets the graphs in Fig. 1(a) for a fixed distance between the slits, and in Fig. 1(b) for a fixed time of flight. A quantum system, characterized by the value  $\mathcal{V} = 1$ , tends to display a classical behavior, characterized by  $\mathcal{V} = 0$ , when the time of flight and/or the distance between the slits are increased. This quantum-classical transition takes place

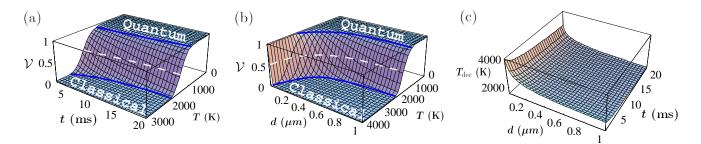


FIG. 1: (a): Visibility  $\mathcal{V}$  vs T (0–3000 K) and t (3–20 ms) for  $d = 1\mu$ m. (b): Visibility  $\mathcal{V}$  vs T (0–4000 K) and d (0.01–1  $\mu$ m) for t = 10 ms. The dashed white level line indicates  $\mathcal{V} = 1/2$  and determines the "decoherence temperature" plotted in (c):  $T_{\text{dec}}$  vs. d (0.02–1  $\mu$ m) and t (1–20 ms). We plot only the physically relevant ranges of the parameters.

at a "decoherence temperature"  $T_{\rm dec}$  determined by the level curve

$$\mathcal{V} = 1/2 \quad \Longleftrightarrow \quad [\Lambda(T) - G(T, d)]t - \ln 2 = 0.$$
 (31)

 $T_{\rm dec}(d,t)$  is plotted in Fig. 1(c). The transition between the quantum and classical behavior is very sharp, both in Figs. 1(a) and 1(b), and this enables us to define the decoherence temperature in a clear-cut way. These graphs are the central results of our analysis.

A mesoscopic system, such as a macromolecule, can be attributed a temperature, in the sense of Eq. (20), by virtue of its large number of freedoms N. In a double slit interference experiment, the degree of freedom associated with the interfering pattern [the relevant variable being x, see Eq. (1)] plays a special role. We now argue that, in general, such an "interfering" freedom is not in the same thermal state as the others.

Let the experiment last for a time t (the time of flight of the molecule in the interferometer) and  $H_{\text{exch}}$  be the interaction Hamiltonian responsible for the coupling between the interfering freedom and the environment. Interference can be observed if

$$\left\langle \int_{0}^{t} H_{\text{exch}} dt \right\rangle_{T} \lesssim \hbar \ll kTt,$$
 (32)

where the average  $\langle \cdots \rangle_T$  is taken over the initial state of the total system [11]. The above one is a condition on the exchanged action and the environmental temperature. For example, if the average in Eq. (32) is understood in the r.m.s. sense and  $\underline{H}_{exch} = -\xi(t)x$ , like in (2), we have  $\langle \int H_{\text{exch}} dt \rangle = \sqrt{\Lambda t} \langle \Delta p \rangle d = \Delta p_{\text{tot}} d \leq \hbar$ where  $\Delta p_{\text{tot}}(T)$  is the total recoil due to a momentum random walk [9]. In such a case the first inequality in (32) is nothing but Heisenberg's inequality and this clarifies the rationale behind it. When this condition is satisfied, the macromolecule interferes. During the interference experiment, energy flows between the environment and the interfering "colder" freedom. Such a freedom, associated with the interfering component  $p_x$ , approaches equilibrium (at a temperature T), via momentum—and energy-transfer, during the momentum random walk

process described above: eventually, visibility vanishes and interference is lost when the first inequality in (32) ceases to be valid. This is a rather fast process, that induces a classical behavior in the (relevant interfering variable of the) mesoscopic system. The thermalization process sets in afterwards, when  $\langle \int H_{\rm exch} dt \rangle \simeq kTt \gg \hbar$ instead of (32), and is much slower. This is, altogether, a remarkable picture, that adds spell to the interfering features of these mesoscopic systems, as well to the many additional problems that must be considered [10] in order to get a complete picture of these phenomena.

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(1999).

[11] We are always neglecting the temperature of the photon bath, see comments after Eq. (1): strictly speaking our environment is made up of two parts (the other freedoms of the molecule + the photon bath), that are not in equilibrium, and T is the "local" temperature of the rotovibrational and electronic modes of the macromolecule, responsible for photon emission. We are not considering the (much) slower equilibration process of the whole environment.