Quantum lithography, entanglement and Heisenberg-limited parameter estimation

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Abstract
We explore the intimate relationship between quantum lithography, Heisenberg-limited parameter estimation and the rate of dynamical evolution of quantum states. We show how both the enhanced accuracy in measurements and the increased resolution in quantum lithography follow from the use of entanglement. Mathematically, the hyper-resolution of quantum lithography appears naturally in the derivation of Heisenberg-limited parameter estimation. We also review recent experiments offering a proof of the principle of quantum lithography, and we address the question of state preparation and the fabrication of suitable photoresists.

Keywords: quantum imaging, quantum metrology, shot-noise, standard quantum limit

An important branch of quantum mechanics is parameter estimation. Heisenberg's uncertainty principle seems to prevent us from determining a physical parameter such as a phase with infinite precision, and it is therefore important to understand what are the limits of the estimation process. It is also a branch of physics that particularly interested Haus (e.g., Haus 1995).

In this paper we consider the link between phase estimation and the rate of dynamical evolution of quantum states. This will lead us to the concept of quantum lithography and the question of the role of entanglement. Finally, we will review some experiments that show the viability of quantum lithography (at least in principle), and briefly consider the generation of the necessary optical quantum states.

1. Parameter estimation

Consider a physical process that induces a relative phase φ in the state of a system. For example, one can send light through a medium with an unknown index of refraction n_r to induce a phase shift $\varphi = 2\pi L(n_r - 1)/\lambda$, with λ the wavelength of the light and L the length of the medium. This phase shift can be measured relative to a reference beam in a Mach–Zehnder

interferometer. When we assume that L and λ are known with very high precision, we can infer the index of refraction with an accuracy that is proportional to the error in the phase. Many precision measurements can be reformulated in terms of a phase measurement, with the search for gravitational waves as one of the most urgent cases (Fritschel *et al* 1998). It is therefore important to have definite bounds on the error in the phase, given a particular set of resources. We should realize, however, that there are two distinct questions we can ask about the uncertainty in the phase.

First, there is the relative phase difference $\delta\varphi$ that renders two quantum states $|\psi(0)\rangle$ and $|\psi(\delta\varphi)\rangle$ distinguishable, i.e.,

$$\langle\psi(0)|\psi(\delta\varphi)\rangle = 0. \tag{1}$$

Here, we assume that the dependency on $\delta\varphi$ actually does render $|\psi(\delta\varphi)\rangle$ orthogonal to $|\psi(0)\rangle$. We can then minimize $\delta\varphi$ to satisfy equation (1). This depends on the characteristics of $|\psi(0)\rangle$ and $|\psi(\delta\varphi)\rangle$. Thus, $\delta\varphi$ is a measure of the dynamical rate of the (unitary) evolution.

A second question we can ask is how well one can measure φ, given a state $|\psi(\varphi)\rangle$. That is, we want an expression for the error $\Delta\varphi$ in the phase. Since there is no phase operator, we

usually measure a suitably chosen (Hermitian) operator $\hat{X}$ such that

$$\Delta\varphi = \frac{\Delta\hat{X}}{|\mathrm{d}\langle\hat{X}\rangle/\mathrm{d}\varphi|}. \tag{2}$$

Mathematically, these two problems are very similar, since measuring $|\psi(\varphi)\rangle$ typically involves interference with some other state $|\psi(\varphi')\rangle$. Choosing situations where $\delta\varphi = \Delta\varphi$ allows us to derive bounds on the fundamental quantum limits of parameter estimation ($\Delta\varphi$) by calculating the maximum rate of dynamical evolution ($\delta\varphi$).

It was recognized early on by Mandelstam and Tamm in 1945 that the uncertainty relation between time and energy for a given frequency ω can be used to prove the following inequality (Mandelstam and Tamm 1945):

$$\Delta\varphi = \omega\Delta t \geqslant \frac{\pi}{2}\frac{\hbar\omega}{\Delta E}, \tag{3}$$

where t is the time it takes to evolve from the initial state to the next orthogonal state, $\hbar\omega$ is the energy difference between the two orthogonal states, and we have used $\delta\varphi = \Delta\varphi$. Furthermore, ΔE is the uncertainty in energy. This inequality can be interpreted as follows: the minimum phase difference $\Delta\varphi$ that can be detected is inversely proportional to the normalized energy spread of the state $|\psi\rangle$ (with $(\Delta E)^2 = \langle\psi|E^2|\psi\rangle - \langle\psi|E|\psi\rangle^2$). The normalization is given by $\hbar\omega$. The factor $\pi/2$ has a geometric interpretation that is highly relevant for this paper: the minimum distance between two points that can be distinguished by a wave is given by the distance between a crest and the adjacent trough of the wave. This (normalized) distance of $\pi/2$ is also known as the *Rayleigh limit*.

However, it became clear that the Mandelstam–Tamm (MT) inequality is not applicable to situations involving certain pathological states (Shapiro *et al* 1989, Braunstein *et al* 1992, Uffink 1993). When coherent (classical) states with $\Delta E = \hbar\omega\sqrt{\langle n\rangle}$ are used, the MT-inequality yields the Poissonian error

$$\Delta\varphi \geqslant \frac{\pi}{2}\frac{1}{\sqrt{\langle n\rangle}}, \tag{4}$$

where $\langle n\rangle$ is the average number of quanta in the coherent state. The indication was that phase estimation could not improve beyond the so-called Heisenberg limit (Ou 1996):

$$\Delta\varphi \gtrsim \frac{1}{\langle n\rangle}. \tag{5}$$

But there exist states with finite average energy E and unbounded ΔE. According to the MT-inequality, such states could have a precision that is proportional to, for example, $\langle n\rangle^{-2}$.

If there are states for which the MT-inequality does not give a saturated bound, then the next question is whether an additional bound can be derived. In 1998, Margolus and Levitin did just that (Margolus and Levitin 1998). They showed that

$$\Delta t \geqslant \frac{\pi}{2}\frac{\hbar}{E}, \tag{6}$$

which for narrow-band schemes around frequency ω with $E = \hbar\omega\langle n\rangle$ yields

$$\Delta\varphi \geqslant \frac{\pi}{2}\frac{1}{\langle n\rangle}. \tag{7}$$

This inequality therefore restricts the precision in a phase measurement by the *average energy*, rather than the energy spread. The pathological states mentioned above turn out to have sufficiently small average energies so that equation (5) is still satisfied. Recent examples of applied phase estimation include frequency measurements (Bollinger *et al* 1996), the quantum gyroscope (Dowling 1998), quantum positioning and clock synchronization (Giovannetti *et al* 2001), and length and weak force measurements using coherent optical states (Ralph 2002, Munro *et al* 2002).

2. The role of entanglement in parameter estimation

Suppose we want to estimate a parameter φ by measuring a particular observable. Construct the state

$$|\varphi\rangle = \frac{1}{\sqrt{2}}\left(|0\rangle + \mathrm{e}^{\mathrm{i}\varphi}|1\rangle\right), \tag{8}$$

where the basis $\{|0\rangle, |1\rangle\}$ is chosen in some convenient way determined by the physics of estimating φ with the choice of observable as $\sigma_x = |0\rangle\langle1| + |1\rangle\langle0|$. Then the expectation value of σ_x is given by $\langle\varphi|\sigma_x|\varphi\rangle = \cos\varphi$. When we repeat this experiment N times, we obtain

$$\langle\sigma_x^N\rangle = {}_1\langle\varphi|\cdots{}_N\langle\varphi|\bigoplus_{k=1}^{N}\sigma_x^{(k)}|\varphi\rangle_1\cdots|\varphi\rangle_N = N\cos\varphi. \tag{9}$$

Furthermore, we know that $\sigma_x^2 = \mathbb{1}$, and the variance of σ_x given N samples is readily computed to be $(\Delta\sigma_x)^2 = N(1 - \cos^2\varphi) = N\sin^2\varphi$. According to estimation theory,

$$\Delta\varphi = \frac{\Delta\sigma_x}{|\mathrm{d}\langle\sigma_x^N\rangle/\mathrm{d}\varphi|}. \tag{10}$$

The standard variance in the parameter φ after N trials is thus given by

$$\Delta\varphi_{\mathrm{st}} = \frac{\sqrt{N}\sin\varphi}{N\sin\varphi} = \frac{1}{\sqrt{N}}. \tag{11}$$

In other words, the uncertainty in the phase is inversely proportional to the square root of the number of trials. This is the classical Poissonian error in the phase.

With the help of quantum entanglement we can achieve the Heisenberg limit of $1/N$. Consider an entangled input state on N systems:

$$|\varphi_N\rangle = \frac{1}{\sqrt{2}}\left(|0, \ldots, 0\rangle_{1\cdots N} + \mathrm{e}^{\mathrm{i}N\varphi}|1, \ldots, 1\rangle_{1\cdots N}\right). \tag{12}$$

The relative phase $\mathrm{e}^{\mathrm{i}N\varphi}$ can be obtained by a unitary evolution $|1\rangle \rightarrow \mathrm{e}^{\mathrm{i}\varphi}|1\rangle$ and $|0\rangle \rightarrow |0\rangle$, thus yielding the required factor. When we suggestively write $|\mathbf{0}\rangle = |0, \ldots, 0\rangle_{1\cdots N}$ and $|\mathbf{1}\rangle = |1, \ldots, 1\rangle_{1\cdots N}$, then the state becomes

$$|\varphi_N\rangle = \frac{1}{\sqrt{2}}\left(|\mathbf{0}\rangle + \mathrm{e}^{\mathrm{i}N\varphi}|\mathbf{1}\rangle\right). \tag{13}$$

This is mathematically equivalent to a single (nonlocal!) system with a relative phase shift of $N\varphi$. In order to measure this phase, we need to measure the (nonlocal) observable Σ_N:

$$\Sigma_N = |\mathbf{0}\rangle\langle\mathbf{1}| + |\mathbf{1}\rangle\langle\mathbf{0}|. \tag{14}$$

This yields

$$\langle\varphi_N|\Sigma_N|\varphi_N\rangle = \cos N\varphi. \qquad (15)$$

As before, we obtain

$$\Delta\varphi_q = \frac{\Delta\Sigma_N}{|\mathrm{d}\langle\Sigma_N\rangle/\mathrm{d}\varphi|} = \frac{1}{N}\frac{\sin N\varphi}{\sin N\varphi} = \frac{1}{N}. \qquad (16)$$

Here we see that the precision in φ is increased by a factor $\sqrt{N}$ over the standard noise limit when we exploit quantum entanglement.

When the loss of subsystems is considered (for example, when n out of N photons fail to arrive), the entangled state in equation (12) becomes separable and mixed:

$$\rho_{N-n} = \tfrac{1}{2}|0\rangle_{1\cdots N-n}\langle 0| + \tfrac{1}{2}|1\rangle_{1\cdots N-n}\langle 1|. \qquad (17)$$

Not only is it separable, there is also no information about φ in this state. This state can therefore not be used for the parameter estimation at all.

One way to circumvent this practical difficulty is to use separable states and nonlocal observables. When $N - n$ out of N systems arrive, the experimenter chooses to measure Σ_{N-n} instead of Σ_N. This way, the parameter φ is estimated with an enhanced precision proportional to $1/(N - n)$. The price to pay is that often the measurement of Σ_{N-n} will fail because it does not span the complete state space. It must also be noted that this trick depends on the physical implementation, and cannot always be applied.

3. Quantum lithography

One particular physical implementation of this process involves light, and this will lead us to the main topic of this paper. Suppose that the state in equation (13) is physically implemented by a so-called two-mode *Noon state* (Kok *et al* 2002):

$$|\varphi_N\rangle = \frac{1}{\sqrt{2}}\left(|N, 0\rangle + \mathrm{e}^{\mathrm{i}N\varphi}|0, N\rangle\right), \qquad (18)$$

where $|k\rangle$ is an k-photon state. When we choose Σ_N to be

$$\Sigma_N = |N, 0\rangle\langle 0, N| + |0, N\rangle\langle N, 0|, \qquad (19)$$

then the uncertainty in the phase is again given by $\Delta\varphi = 1/N$. However, there is also something else going on ….

When we calculate the expectation value of Σ_N, we find that it varies as $\cos N\varphi$. That is, the geometric distance between the two points that can be distinguished according to the Rayleigh limit is now N *times smaller than in the classical case*! As a consequence, we can in principle read and write much smaller features with this technique. This is called quantum lithography (Boto *et al* 2000), and we will now give a full description of this phenomenon in terms of the quantum interference of two optical modes.

3.1. Interference on a surface and the Rayleigh limit

Suppose two plane waves characterized by $\vec{k}_1$ and $\vec{k}_2$ hit a surface at an angle θ from the normal vector. The wavevectors are given by

$$\vec{k}_1 = k(\cos\theta, \sin\theta) \qquad \text{and} \qquad \vec{k}_2 = k(\cos\theta, -\sin\theta), \qquad (20)$$

where we assume $|\vec{k}_1| = |\vec{k}_2| = k$. The wavenumber k is related to the wavelength of the light according to $k = 2\pi/\lambda$.

In order to find the interference pattern in the intensity I, we sum the two plane waves at position $\vec{r}$ at the amplitude level:

$$I(\vec{r}) \propto \left|\mathrm{e}^{\mathrm{i}\vec{k}_1\cdot\vec{r}} + \mathrm{e}^{\mathrm{i}\vec{k}_2\cdot\vec{r}}\right|^2 = 4\cos^2\left[\tfrac{1}{2}(\vec{k}_1 - \vec{k}_2)\cdot\vec{r}\right]. \qquad (21)$$

When we calculate the inner product $(\vec{k}_1 - \vec{k}_2)\cdot\vec{r}/2$ from equation (20) we obtain the expression

$$I(x) \propto \cos^2(kx\sin\theta) \qquad (22)$$

for the intensity along the substrate in direction x.

As we saw above, the Rayleigh limit is given by the minimal resolvable feature size Δx that corresponds to the distance between an intensity maximum and an adjacent minimum. From equation (22) we obtain

$$k\Delta x\sin\theta = \frac{\pi}{2}. \qquad (23)$$

This means that the maximum resolution is given by

$$\Delta x = \frac{\pi}{2k\sin\theta} = \frac{\pi}{2\left(\frac{2\pi}{\lambda}\sin\theta\right)} = \frac{\lambda}{4\sin\theta}. \qquad (24)$$

The maximum resolution is therefore proportional to the wavelength and inversely proportional to the sine of the angle between the incoming plane waves and the normal. The resolution is thus maximal (Δx is minimal) when $\sin\theta = 1$, or $\theta = \pi/2$. This is the limit of grazing incidence. The classical diffraction limit is therefore $\Delta x = \lambda/4$. Note that this derivation does not use the approximation $\sin\theta \simeq \theta$, which is common when considering diffraction phenomena.

3.2. Surpassing Rayleigh's diffraction limit

So how does quantum lithography work? In the limit of grazing incidence, we let the two counterpropagating light beams a and b be in the combined entangled state of N photons

$$|\psi_N\rangle_{ab} = \left(|N, 0\rangle_{ab} + \mathrm{e}^{\mathrm{i}N\varphi}|0, N\rangle_{ab}\right)/\sqrt{2}, \qquad (25)$$

where $\varphi = kx$, with $k = 2\pi/\lambda$. We define the mode operator $\hat{e} = (\hat{a} + \hat{b})/\sqrt{2}$ and its adjoint $\hat{e}^\dagger = (\hat{a}^\dagger + \hat{b}^\dagger)/\sqrt{2}$. The deposition rate Δ on the substrate is then given by (Boto *et al* 2000)

$$\Delta_N = \langle\psi_N|\hat{\delta}_N|\psi_N\rangle \qquad \text{with } \hat{\delta}_N = \frac{(\hat{e}^\dagger)^N\hat{e}^N}{N!}, \qquad (26)$$

i.e., we look at the higher-order moments of the electric field operator on the substrate. The deposition rate Δ scales with the Nth power of intensity. Leaving the substrate exposed for a time t to the light source will result in an exposure pattern $P(\varphi) = \Delta_N t$. After a straightforward calculation we see that

$$\Delta_N \propto (1 + \cos N\varphi). \qquad (27)$$

We interpret this as follows. A path-differential phase-shift φ in light beam b results in a displacement x of the interference

pattern on the substrate. Using two classical waves, a phase-shift of 2π will return the pattern to its original position. However, according to equation (27), one cycle is completed after a shift of $2\pi/N$. This means that a shift of 2π will displace the pattern N times. In other words, we have N times as many maxima in the interference pattern. These will be closely spaced, yielding an effective Rayleigh resolution of $\Delta x = \lambda/(4N)$, a factor of N below the classical interferometric result of $\Delta x = \lambda/4$.

We have also shown that quantum lithography can be used to create arbitrary patterns in one dimension, rather than just closely spaced lines (Kok *et al* 2001). This can be achieved by using superpositions of the more complicated state

$$|\psi_{Nm}\rangle = \frac{1}{\sqrt{2}}\left(e^{im\varphi}|N-m,m\rangle + e^{i(N-m)\varphi}|m,N-m\rangle\right).$$

(28)

There are two fundamentally different ways states of the form in equation (28) can be superposed. We can either sum over the photon number N:

$$|\Psi_m\rangle = \sum_{n=0}^{N} \alpha_n |\psi_{nm}\rangle,$$

(29)

with α_n complex coefficients, or we can sum over the photon distribution m:

$$|\Psi_N\rangle = \sum_{m=0}^{\lfloor N/2 \rfloor} \alpha_m |\psi_{Nm}\rangle,$$

(30)

where $\lfloor N/2 \rfloor$ denotes the largest integer l with $l \leqslant N/2$ and α_m again the complex coefficients. Every branch in this superposition is an N-photon state.

These techniques not only allow us to create arbitrary one-dimensional patterns, but extensions to four-mode states also facilitate two-dimensional patterns (Kok *et al* 2001). Furthermore, by choosing the α_m coefficients of equation (30) in a special way, Björk *et al* (2001) showed that one can construct a subwavelength resolution pixel state. This state can subsequently be used to illuminate a surface in order to etch an arbitrary pattern.

3.3. Demonstration of quantum lithography and the preparation of states

The principle of quantum lithography has been demonstrated using two-photon path-entangled states generated by parametric down-conversion (d'Angelo *et al* 2001). In this experiment, the photon pairs are created in a BBO crystal (β-BaB_2O_4). Immediately behind the crystal, a double-slit aperture is placed, which blocks most pairs. However, when two photons do get through, they are extremely likely to have passed through the same slit.

This way, the state of the light field just after the slits is approximately $|2,0\rangle_{AB} + |0,2\rangle_{AB}$, where A and B denote the two slits. This state is of the form of equation (25), and can therefore be used to beat the Rayleigh limit. By scanning the output field with a set of two detectors, the diffraction pattern conditioned on a twofold detector coincidence was mapped out. This pattern was twice as narrow as the single-photon diffraction pattern, and the principle of sub-Rayleigh resolution due to two-photon quantum lithography was thereby

demonstrated. Subsequent experiments with photon pairs have confirmed these findings (Edamatsu *et al* 2002, Shimizu *et al* 2002).

However, in order for quantum lithography to work, we also need a suitable photoresist. In other words, we need a material that is sensitive to multi-photon events in order to etch images on its surface. Last year, Korobkin and Yablonovitch (2002) exposed commercial photographic film to photon pairs, and produced the coveted twofold resolution enhancement.

Shortly after the experiment of d'Angelo *et al*, Cataliotti *et al* (2001) performed frequency measurements using multi-photon Raman transitions in rubidium atoms that are confined in an optical dipole trap. The multi-photon events of up to fifty photons resulted in a spectral width that is below the Fourier limit. Although not exactly quantum lithography, this experiment strongly suggests that the *quantum Rayleigh limit* of $\lambda/(4N)$ is correct.

Another essential ingredient for quantum lithography is the generation of the required quantum states of the light field. The states in equations (28) and (29) are very complicated, and it is not quite clear how they can be generated efficiently without large Kerr nonlinearities (Gerry and Campos 2001). The production of N-photon entangled states conditioned on *non*-detection was proposed (Fiurášek 2002), as well as the creation of Noon-states based on single-photon detection (Lee *et al* 2002, Kok *et al* 2002, Gerry *et al* 2002).

4. Conclusions

We have shown that quantum lithography and Heisenberg-limited parameter estimation are two manifestations of the same principle: instead of many separate measurements of φ that lead to the shot-noise limit (in, for example, an experiment using N trials with the single-photon path-entangled state $|1,0\rangle + e^{i\varphi}|0,1\rangle$), entangling the resources of these N measurements to conduct a single-shot experiment (e.g., using the N-photon Noon-state $|N,0\rangle + |0,N\rangle$) can reduce the noise to the Heisenberg limit. Similarly, instead of Rayleigh-limited single-photon diffraction patterns, we can use N-photon entangled states to increase the resolution by a factor N. Mathematically, the hyper-resolution of quantum lithography appears naturally in the derivation of Heisenberg-limited parameter estimation.

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