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CORRELATION HOLOGRAPHY:

IMAGING OF ATOMS WHEN $\sigma_{\text{inelastic}} \gg \sigma_{\text{elastic}}$

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~~Abstract~~

Atomic-scale resolution of details is possible with this method, even if photons interact with the atoms overwhelmingly inelastically, i.e. when $\sigma_{\text{inelastic}} \gg \sigma_{\text{elastic}}$. Observation of small objects is compatible with quantum mechanics even if the disturbance of the object caused by the observation process is arbitrarily small.

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High resolution holograms would be of great value to many branches of science including biology.

It has been argued repeatedly¹ that atomic-scale holographic resolution will not be possible for atoms of most interest in biology, because for these atoms the cross section for elastic photon interactions, σ_e , is much smaller than the one for inelastic² photon interactions, σ_i , so that $\sigma_i / \sigma_e \gg 1$ whenever the photon wavelength, λ , is in the interesting range. The essence of the argument is as follows: To see the holographic image of an atom, a photon has to be coherently scattered off the atom. Since $R \gg 1$, the probability for such scattering is much less than the probability that the photon is absorbed. We know that photon absorption leads with a high probability to the dissociation of molecules. Therefore, the molecule will dissociate almost certainly before a holographic picture of it can be taken. The most one can hope for by observing the points of impact of photons in the hologram is to obtain very limited information about the molecule (e.g. the position of only some of its atoms, its projections along one axis, etc.) according to conventional wisdom.

My purpose here is to point out that the relation $R \gg 1$ is not sufficient to disprove the possibility of atomic-scale-resolution holograms of biological molecules: If photons were Boltzmann particles, then the reasoning just outlined would be compelling. But photons obey Bose-Einstein statistics, thus certain correlations exist between photons which are absent between Boltzmann particles. These correlations can be exploited to make imaging possible.

Suppose that we wish to produce an image of a molecule containing many atoms, i.e. record the position of the center of mass of the atoms relative to each other. Let us fix our attention on one of the atoms, located at point P_0 . Illuminate it with an "incoming" monochromatic electromagnetic plane wave radiation (e.g., X-rays) of energy ω , momentum $\hat{k}$ and amplitude A_1 .

The atom scatters this wave, the elastic scattered amplitude is A_s .

The number of incoming photons crossing a unit surface at normal incidence per unit time is $I_i = \alpha |A_i|^2$, where α is a normalization constant. Similarly, the photon flux density in the scattered wave is $I_s = \alpha |A_s|^2$.

The final ("outgoing") amplitude of photons with energy ω is, as a result of scattering by the atom, $A_f = A_i + A_s$. The final flux density of photons with energy ω is

$$I_f = \alpha |A_f|^2 = \alpha |A_i|^2 + \alpha 2 \operatorname{Re} (A_i^* A_s) + \alpha |A_s|^2. \quad (1)$$

when $|A_s| \ll |A_i|$, as is expected, the last term is negligible, so that the variation in flux density of photons with energy ω , due to scattering by the atom is

$$\Delta I = I_f - I_i \approx \alpha 2 \operatorname{Re} (A_i^* A_s). \quad (2)$$

This is proportional to A_i^* for fixed A_s , therefore, it can be arbitrarily large, even if the atom is likely to be destroyed before it has a chance to elastically scatter even one photon.

So far we showed that an arbitrarily intense photon flux density signal can be generated by the atom. It remains to be seen whether the signal can be distinguished from the background "noise" flux density I_i .

The average of the number, N_i , of incoming photons impinging normally on a surface area S during a time interval T , is $ST \propto |A_i|^2$. The probable deviation of N_i from its average value $\bar{N}_i$ is $\sim$ ("proportional to") $N_i^{1/2} \sim A_i$. Since $A_s \sim A_i$, one has $\Delta I \sim |A_i|^2 \sim |A_s|^2$, so that the signal to noise ratio is $\sim |A_s|$. (The constant background N_i can always be subtracted.)

How large an A_s does one need for the signal to be observable? Assume that the detector consists of several identical sized detecting surfaces. The signal generated on the j^{th} surface will exceed the background fluctuation there, if

$$|ST \propto 2|A_i||A_s|\cos\theta_j \geq (ST \propto |A_i|^2)^{1/2}, \quad (3)$$

where θ_j is the phase angle between A_i and A_s on that surface (provided the angle of incidence of all waves is close to $\frac{1}{2}\pi$ there). Summing over all detector surfaces (i.e. summing over θ_j) one finds from Eq. (3) that the signal will be distinguishable from the background if $ST \propto |A_s|^2 \geq 1$. But the L.H.S. of this inequality is just $STI_s = N_s$, the number of scattered photons impinging on the detector. Thus one needs

$$N_s \geq 1. \quad (4)$$

The total number, N_{st} , of photons scattered by the atom during T cannot be less than N_s (since some scattered photons may not reach the surface). Therefore, even under the best of circumstances (when the detector surface completely encloses the atom) the presence of the atom cannot be detected by observing the interference term, ΔI , unless at least one photon has been scattered elastically by the atom. But chances for an elastic photon scattering are R^{-1} , negligible³ since by assumption $R \gg 1$.

The validity of inequality (4) is more general. Suppose that one wishes to observe a number, N^B , of Boltzmann particles impinging on a surface, on which a random "background" consisting of N_i particles of the same kind is also incident. Suppose further that the measurement is made by recording the points of impact of all the particles reaching the surface. Then whenever N^B can be written as $N^B = N_{st}^{1/2} N_i^{1/2}$, the condition insuring that N^B will be observable against the background fluctuation is just condition (4).

Photons, however, are not Boltzmann particles. The location and time of arrival of one photon is correlated with those of other photons, as demonstrated, e.g. by R. Hanbury Brown and R.Q. Twiss⁴. For the steady state situation considered here, time correlations are uninteresting, but the spatial ones can be

observed in addition to the photon points of impact, and may serve to distinguish the signal from the background noise.

To illustrate, assume that we wish to image a single molecule, with typical diameter, d . The molecular center of mass is located at P_0 . Let $\vec{r}_0$ be a radius vector measured from P_0 to some point P . The scattered amplitude, A_s , will be a wave diverging from P_0 . The interference between A_s and A_i will produce a pattern which at any point P far from P_0 will vary (almost) harmonically⁵ along $\vec{r}_0$, with a wavelength $\Lambda = \lambda (1 - \cos \Theta_0)^{-1}$, where Θ_0 is the angle between $\vec{k}_i$ and $\vec{r}_0$ (see Figure 1). Since the molecule is finite, the A_s will be a superposition of terms, each originating at a different point P_ℓ within the molecule. Therefore, the interference pattern will contain a sum of terms, each varying with a slightly different $\Lambda_\ell = \lambda (1 - \cos \Theta_\ell)^{-1}$, where Θ_ℓ is the angle between $\vec{k}_i$ and $\vec{r}_\ell$, and $\vec{r}_\ell$ is the radius vector from P_ℓ to P . When P is far from the molecule, all Λ_ℓ will lie within the range $\Lambda_0 (1 \pm \frac{1}{2} \Delta \Lambda_0)$ where, for small Θ_0 , $\Delta \Lambda_0 = 4d/r_0 \Theta_0$, decreasing as $|\vec{r}_0| d^{-1} = r_0 d^{-1}$ increases.

Suppose now that the photosensitive medium (e.g. photographic film) is not placed only on the surface S , but is diluted to fill a volume between S and the molecule (delineated by dashed lines in Fig. 1). The three-dimensional interference pattern between A_i and A_s can then be recorded. Taking a slice of this pattern along any chosen $\vec{r}$, far from P_0 , the signal of interest as a function of r will have Fourier components with wavelength in the region $\Lambda_0 (1 \pm \frac{1}{2} \Delta \Lambda_0)$. On the other hand, the background fluctuation due to $|A_i|^2$ will vary randomly⁶ as a function of r . Let m be the number of intervals Δr along $\vec{r}$ to be resolved in the slice under study. If, as is likely, $m < \Lambda_0 / \Delta \Lambda_0$, then one can distinguish m different Fourier components as a function of r . Only a fraction $\frac{1}{m}$ of the background is likely to have a variation with Fourier components in the interval $\Lambda_0 (1 \pm \frac{1}{2} \Delta \Lambda_0)$, thus the

term ΔI due to elastic photon scattering by an atom in the molecule will not be observable over the background, unless the number of scattered photons reaching the slice satisfies instead of condition (4):

$$N_s \gtrsim \frac{1}{m}. \quad (5)$$

When the recording surface (and medium) has detection efficiency f (because of incomplete solid angle coverage, etc.), then the atom will be detectable provided that the total number of elastically scattered photons by the atom is $N_{st} \gtrsim m^{-1} f^{-1}$, i.e. for large m the presence of the atom can be detected by elastic photon scattering even if the number of expected photons scattered elastically by the atom is much less than one.

Next we discuss the solutions of two problems which are important for a practical application of the above considerations.

First, if one wishes to image an object with resolution δ , then by the Heisenberg uncertainty relation one needs photons momentum components up to $\hbar/\delta$ along the surface of the object. If $\vec{k}_1$ is perpendicular to the surface, this requirement states that the imaging process must make use of photons scattered at angles up to $\Theta = \arcsin(\lambda/\delta)$. Then in the method here proposed, one will have to observe interference patterns with

$\Lambda \gtrsim \lambda \left[1 - (1 - \lambda^2/\delta^2)^{1/2} \right]^{-1}$. When $\lambda \approx \delta$, then $\Lambda \approx \lambda$, and if δ is of the order of an atomic radius, one would need a detector capable of recording with this resolution. Such recorders are not known. A sufficient interference pattern can nevertheless be recorded⁷ as follows: Record along r the interference between A_s and (not A_i but) A_i' , the amplitude of a wave with energy ω momentum $\vec{k}_i'$ travelling at an angle $\Theta' \ll 1$ with respect to $\vec{r}$ (Fig 1).

Thus the photons can still be scattered up to large angles Θ by the molecule, but they interfere with a wave which travels almost parallel to them, so that $\Lambda = \lambda (1 - \cos \Theta') \gg \lambda$, and can be recorded. In practice, A_i' may be a spherical wave diverging from a point P' , whose distance from P_0 is $\ll$

the distance where the pattern is observed: $|\vec{r}'| \ll |\vec{r}|$. Furthermore, it seems to have been overlooked that the Λ can be made larger when λ is decreased, since if λ is small, $\theta \approx \arcsin(\lambda/\delta) \approx \lambda/\delta \ll 1$, suffices, and then $\Lambda \sim \lambda/\theta^2 \sim \lambda^{-1}$.

Second, the recorded interference pattern can be analyzed by counting grain density as a function of $\vec{r}$ (if the medium is a photographic film). A more elegant procedure is based on the recognition that any thin slice of the recording medium in which the originally recorded grain density (and with proper film development) the light amplitude transmissivity of the developed film varies harmonically with wavelength Λ , will act as a harmonic potential: It will allow part of the incident photons to pass without change of momentum, but will transfer momentum h/Λ to the rest, i.e. deflect them parallel (or antiparallel) to $\vec{r}$ by an angle $\arctan(\lambda/\Lambda)$. Fourier analysis of any slice can be accomplished by simply observing the amplitude of transmitted light as a function of angle (even for smaller r , when the signal variation is not simple harmonic).

One may argue that the method here described is wrong in principle: To observe an interference pattern produced by an atom, there must be many photons scattered by the atom (e.g. many silver grains blackened). But that is impossible if $\sigma_{\text{elastic}}/\sigma_{\text{elastic}} \ll 1$. The fallacy in this argument is the phrase "photons scattered by the atom." Quantum mechanics does not allow one to specify that the photons came from the atom, no more than it would allow one to assume that photos producing an interference pattern behind a slit grating passed through any one particular slit. One can only say that any interfering photon has partly (quite weakly in the present instance) interacted with the atom, and partly bypassed it in the quantum mechanical sense. The energy flow needed to e.g. blacken the silver grains in a photographic film is provided in our case primarily by $|A_1|^2$; the small scattered amplitude A_s serves mainly to regulate where it is deposited.

We conclude that it is possible to establish experimentally the presence of an atom (or an array of atoms), by observing the amplitude A_s of scattered photons, even if A_s is so low that it corresponds to a time integrated flux of scattered photons which is less than one, in fact arbitrarily small. It is possible to image a molecule by observing the interference pattern of photons scattered elastically by that molecule, even when $\sigma_{\text{inelastic}} / \sigma_{\text{elastic}} \gg 1$. It is compatible with the principles of quantum mechanics to clearly observe an object, even if chances are negligible that the observation will in any way disturb the object⁸.

References, Footnotes

1. For example, J.R. Breedlove, G.T. Trammell, *Science* 170, 1310 (1970); G.T. Trammell, *Physics Today* 57, (July 1976); G. Chapline and L. Wood, *Physics Today* 51 (July 1976).
2. The inelastic processes are mainly photoelectric. Compton scattering and other inelastic reactions are less important in the region of interest.
3. One can overcome this difficulty in certain cases by embedding many molecules of the same kind in a crystal, to coherently enhance A_s . This is done in X-ray diffraction studies. Unfortunately, most molecules cannot be so embedded.
4. R. Hanbury Brown and R.Q. Twiss, *Nature* 177, 27, and 178, 1447.
5. Superimposed on a fast variation with wavelength $\lambda / (1 + \cos \theta_s)^{-1}$, which is generally unobservable for small λ .
6. Except for a known attenuation due to absorption in the medium.
7. Paul L. Csonka, *Particle Accelerators* 8, 161 (1978), and in "Wiggler Magnets," Ed. H. Winick and T. Knight, p. II.162, SSRP Report 77/05 (May 1977).
8. The uncertainty principle is clearly not violated; photon position and momentum are measured, but not simultaneously, of course.

Figure Caption

The center of mass of a molecule of typical dimension d is located at P_0 . It is illuminated by a monochromatic electromagnetic wave with momentum $\vec{k}_i$ and an amplitude A_i . The wave scattered by the molecule has amplitude A_s . The interference between A_s and A_i (or A_i') is recorded in the photosensitive medium delineated by the surface S and the dashed lines. (Other symbols defined in the main text.)