



Testing quantum mechanics in non-Minkowski space-time with high power lasers and 4th generation light sources

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A common misperception of quantum gravity is that it requires accessing energies up to the Planck scale of 10^{19} GeV, which is unattainable from any conceivable particle collider. Thanks to the development of ultra-high intensity optical lasers, very large accelerations can be now the reached at their focal spot, thus mimicking, by virtue of the equivalence principle, a non Minkowski space-time. Here we derive a semiclassical extension of quantum mechanics that applies to different metrics, but under the assumption of weak gravity. We use our results to show that Thomson scattering of photons by uniformly accelerated electrons predicts an observable effect depending upon acceleration and local metric. In the laboratory frame, a broadening of the Thomson scattered x ray light from a fourth generation light source can be used to detect the modification of the metric associated to electrons accelerated in the field of a high power optical laser.

To date, one of the best examples of weak gravity modifications of quantum mechanics is the experiment of Colella, Overhauser and Werner^{1,2}. In that experiment, the interference pattern of a beam of neutrons, split into two legs that travel at different heights in the Earth's gravitational field, was observed. Another possible observable effect that has been discussed in the literature is the decoherence of matter waves through coupling to a fluctuating metric^{3,4}. To obtain a consistent description of experimental findings, it was shown that the Hamiltonian must consist of two parts, the usual kinetic term and a gravitational term. This forms the basis of a sub-class of semi-classical Einstein equations, known as Newton-Schrödinger Hamiltonians^{5,6}.

Here we follow a simplified approach and take the metric tensor to be of the isotropic form: $g_{\mu\nu} = \text{diag}(h_0^2, -h_1^2, -h_1^2, -h_1^2)$. Under weak gravity, $g_{\mu\nu} = \eta_{\mu\nu} + \delta g_{\mu\nu}$, where $\eta_{\mu\nu}$ is the Minkowski metric of flat space and $\delta g_{\mu\nu}(\phi)$ is the perturbation given in terms of the potential ϕ . In our model, the metric is assumed static. We need then to address the quantum mechanical treatment of the electron. The approach we use is to generalize the standard Hilbert-Schrödinger formalism to curved space-time represented in terms of a metric that depends continuously on the coordinates. In other words, we do not concern ourselves with the quantized nature of the gravitational field itself. This semiclassical approach seems reasonable for weak gravity due to an external source that is not coupled to the motion of the particles under consideration. The generalization of the Klein Gordon equation to curved space-time is done by introducing the covariant derivatives in the Laplacian operator⁷:

$$\left(\frac{1}{\sqrt{g}} \partial_\mu \sqrt{g} g^{\mu\nu} \partial_\nu + \frac{m^2 c^2}{\hbar^2} \right) \psi = 0, \quad (1)$$

where $g = -\det(g_{\mu\nu})$, m is the electron mass, c the speed of light and ψ the electron wavefunction. Upon promoting the metric to an operator status, we construct the Hermitian covariant momentum operator⁸ as $p_\mu = (-i\hbar/g^{1/4})\partial_\mu g^{1/4}$. Extracting the $-p_0 c$ term from (1) yields the non-relativistic Hamiltonian as



$$\mathcal{H} = h_0 mc^2 - \frac{1}{2m} \frac{h_0^{1/2}}{\sqrt{g}} p_a \sqrt{g} g^{ab} p_b \frac{h_0^{1/2}}{\sqrt{g}}, \quad (2)$$

where latin indices are used for the spatial components (with $\mathbf{p} \equiv p_a$), while greek letters are used for 4-vectors. Finally the effect of an external electromagnetic field (*i.e.*, the probe field in a Thomson scattering experiment) is introduced into the Hamiltonian (2) via a gauge transformation $\mathbf{p} \rightarrow \mathbf{p} - e\mathbf{A}$, where e is the electron charge.

The specific form of the coefficients h_0 and h_1 depends on the chosen metric, and limited by the requirement that the Ricci curvature should be zero or second order in the acceleration. In the weak gravity limit we can expand the metric coefficients in terms of exponentials, such that $h_0 = e^\phi$ and $h_1 = e^{\xi\phi}$, where the Rindler metric¹ applies for $\xi = 0$, and the Schwarzschild metric for $\xi = -1$. The exponential factors in the metric coefficients are associated with a localized gravity. In order to emulate a gravitational-like field in the laboratory we use the principle of conformal equivalence whereby the system (electron plus probe field) is equivalently described by a space-time metric which yields the same dynamical behavior. In a terrestrial laboratory, such a metric should not affect the electromagnetic field and the only non-trivial metric with this property is the variable mass metric ($h_0 = h_1$).

Results

Let's consider the case of an electron oscillating in the field produced by the superposition of two laser beams with orthogonal polarizations, with vector potential $\mathbf{A} = \mathbf{A}_0 + \mathbf{A}_1$, and $\mathbf{A}_0 \cdot \mathbf{A}_1 = 0$. We take \mathbf{A}_0 to be a strong low-frequency optical laser field in which the motion of the electron can be treated classically, such that $|e\mathbf{A}_0| \gg (\hbar m \omega_0^3)^{1/2}$, with ω_0 the field frequency. Let the field component \mathbf{A}_1 be a relatively weak perturbative high frequency (x ray) laser field, the effects of which on the electron are treated quantum-mechanically. Hence, $|e\mathbf{A}_1| \ll (\hbar m \omega_1^3)^{1/2}$, with $\omega_1 \gg \omega_0$. We can decompose the canonical momentum as $(\mathbf{p} - e\mathbf{A})^2 \sim (\mathbf{p}_1 - e\mathbf{A}_1)^2 + (\gamma_0 mc v_0)^2$, where $\gamma_0 = (1 - v_0^2/c^2)^{-1/2}$, and $v_0 = |\mathbf{v}_0|$ is the velocity of the electron due to the optical laser field only, which yields that the energy of the electron as $E = [(\mathbf{p}_1 - e\mathbf{A}_1)^2 c^2 + \gamma_0^2 m^2 c^4]^{1/2}$. Replacing γ_0 with a function of the space-time coordinates alone puts this equation in a form that is conform equivalent to a general relativistic Hamiltonian with metric given by $h_0 = h_1 = \gamma_0 = e^\phi$, with acceleration $\mathbf{a} = \dot{\mathbf{v}}_0 = c^2 \gamma_0^{-2} \nabla \phi$. This transformation can be achieved for a single electron by replacing γ_0 with the first integral of the equation of motion in the field \mathbf{A}_0 . What that we have obtained in this way corresponds to the variable mass metric model ($\xi = 1$), so called because the Hamiltonian form for the energy is the same as that for a particle in Minkowski space-time whose mass varies proportionally to $h_0 = h_1$. For a system of many electrons, we assume that the effective extent of the system is small compared with the amplitude of the motion. This means either the electrons occupy a small region or the probed region is small. The underlying idea that dynamical processes confined to one (space) dimension can affect the other dimensions via a distortion of the metric is not new and has appeared in extended versions of general relativity such as Kaluza-Klein^{9,10}.

Scattering of x-ray photons by electrons accelerated in the field of an optical laser. Our goal is to design an experiment where it may be possible to test some aspects of general relativity in the laboratory by emulating the variable mass metric through the acceleration of electrons in an intense optical laser field (which corresponds to the field \mathbf{A}_0), and by probing them by scattering of x ray photons (given by the field \mathbf{A}_1). Thanks to the development of chirped pulse amplification of optical laser light¹¹, state-of-art laser systems can now achieve focused intensities $I \lesssim 10^{20}$ W/cm², thus being able to accelerate the electron to unprecedented values:

$$a = \left| \frac{e\dot{\mathbf{A}}_0}{m} \right| = 4.8 \times 10^{16} \text{I}^{1/2} \text{ cm/s}^2 \quad (3)$$

This poses as an ideal tool to study weak gravity models¹²⁻¹⁴. For the field \mathbf{A}_1 let's take instead the one generated with a fourth generation source, or Free Electron Laser (FEL)¹⁵. Without loss of generality, for the rest of the paper we will replace \mathbf{p}_1 by \mathbf{p} , and \mathbf{A}_1 by \mathbf{A} .

For the present, we assume that the scattering takes place on a sufficiently short timescale ($t \lesssim c/a$) and is confined to a region of FEL spot in which the acceleration field is relatively homogeneous. Accordingly, a can be considered to be constant. More importantly, in an accelerated frame, the conservation of particles does not always hold. Indeed, the most prominent manifestation of quantum gravity is that black holes radiate energy at the universal temperature - the Hawking temperature¹⁶. This is a quite general fact, not confined to black holes. As shown by Davies, Unruh and Fulling¹⁷⁻¹⁹, an observer in a uniformly accelerated frame experiences the surrounding vacuum as filled with thermal radiation with temperature $T_{DU} = \hbar a / 2\pi k_B c = 4.05 \times 10^{-23} a$ K (k_B is the Boltzmann constant). However, when $mc^2, \hbar\omega_1 \gg k_B T_{DU}$, the change in the number of electrons and x ray photons will be negligible (a restatement of the fact that we are still in the weak field limit, well below the Schwinger limit).

As it is customary, we regard the scattering part of the Hamiltonian to be determined solely by the $\mathbf{A}^\dagger \cdot \mathbf{A}$ term. Indeed, for an ensemble of weakly-coupled free electrons (*e.g.*, an ideal electron gas) the polarization contribution from second order terms involving $\mathbf{p} \cdot \mathbf{A}$ terms can be neglected. This is particularly true to the scattering of x rays if the FEL photon energy is much higher than the plasma frequency of the electron gas²⁰. Thus the effective interaction Hamiltonian for the scattering of radiation from an ensemble of electrons in a local gravitational field and confined in a volume $V = \int \sqrt{\gamma} d^3x$ is

$$\mathcal{H}' = \frac{e^2}{2m} \int_V e^{-\phi} \mathbf{A}^\dagger(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) \rho(\mathbf{r}) \sqrt{\gamma} d^3x, \quad (4)$$

where $\rho(\mathbf{r})$ is the electron density operator, and $\sqrt{\gamma} = \sqrt{-\det(g_{ab})} = e^{3\phi}$ is the Jacobian of the induced metric on the 3-dimensional spatial manifold. We use a second quantized representation for the electromagnetic field,

$$\mathbf{A}(\mathbf{r}) = -i \sqrt{\frac{\hbar}{V \epsilon_0}} \sum_{\mathbf{k}, \epsilon} \frac{e e^{i\mathbf{k} \cdot \mathbf{r}}}{\sqrt{\omega}} \mathbf{a}_{\mathbf{k}, \epsilon}, \quad (5)$$

where $\mathbf{a}_{\mathbf{k}, \epsilon}$ is the annihilation operator for a photon in a state of wavenumber \mathbf{k} , frequency ω , polarization ϵ , and ϵ_0 is the vacuum permittivity. Such a representation follows directly from Eq. (1) with the variable mass metric when $m = 0$. Since the variable mass metric only affects particles with a finite rest mass, photons are unaffected by it. We note that in the weak gravity limit, when pair creation is negligible, the vacuum state remains unchanged, and the Bogolubov transformation is not required. Consequently the standard representation of the electromagnetic field in Minkowski space-time is applicable.

Calculation of the differential cross section. The double differential scattering cross section for the x ray photons is given in the Born approximation as:

$$\frac{d^2 \sigma(q, \omega)}{d\omega_f d\Omega} = \frac{V}{c} \mathcal{D} \sum_f \left[\langle \langle \beta; \mathbf{k}_i, \epsilon_i | \mathcal{H}'(t/2) | \alpha; \mathbf{k}_f, \epsilon_f \rangle \rangle \right. \\ \left. \times \langle \langle \alpha; \mathbf{k}_f, \epsilon_f | \mathcal{H}'(-t/2) | \beta; \mathbf{k}_i, \epsilon_i \rangle \rangle \right] dt, \quad (6)$$

where β (α) refers to the initial (final) electron state, $\omega = \omega_i - \omega_f$ is the change in x ray photon frequency due to scattering, and $\mathbf{q} = \mathbf{k}_i - \mathbf{k}_f$ the corresponding change in photon wavenumber, ϵ_i (ϵ_f) the



initial (final) polarization, and $D = V\omega_f^2 / 8\pi^3 c^3$ is the density of final states. N is the total number of electrons in the scattering volume. We then obtain (see supplementary information):

$$\frac{d^2\sigma(\mathbf{q}, \omega)}{d\omega_f d\Omega} = Nr_e^2 (\epsilon_f \cdot \epsilon_i) \frac{\omega_f}{\omega_i} S(\mathbf{q} + i\lambda\mathbf{a}/c^2, \omega), \quad (7)$$

where r_e is the classical electron radius, and $S(\mathbf{k} = \mathbf{q} + i\lambda\mathbf{a}/c^2, \omega)$ is the density-density (Van Hove) correlation function²¹, for particles undergoing collective acceleration \mathbf{a} . The coefficient λ depends on the choice of the metric, and for the variable mass metric defined above yields $\lambda = 2$ (see supplementary information). Eq. (7) is formally identical to the formula describing the scattering of photons from an ensemble of electrons in the absence of a gravitational (or acceleration) field²¹. The effect of acceleration, however, is to introduce an imaginary component into the scattering wavenumber. The density-density correlation (or dynamical form factor) function is given by²¹

$$S(\mathbf{k}, \omega) = \frac{1}{2\pi N} \int_{-\infty}^{\infty} \langle \rho_{-\mathbf{k}}^\dagger(t/2) \rho_{-\mathbf{k}}(-t/2) \rangle e^{i\omega t} dt, \quad (8)$$

where $\rho_{\mathbf{k}}(t) = \sum_{j=1}^N e^{i\mathbf{k}\cdot\mathbf{r}_j(t)}$, is the spatial Fourier transform of the density operator, and $\langle \dots \rangle$ denotes a time average over the scattering process. We also note that Eq. (8) is written in the less common time-symmetric form²¹, which is more appropriate to non-equilibrium systems. This preserves the reality of the density-density correlation function for any value of \mathbf{k} and ω , which is an essential requirement since the differential cross section represents an experimental observable (*i.e.*, the number of scattered photons).

The next step in the evaluation of Eq. (8) is to write the position operator for the j -th electron as $\mathbf{r}_j(t) = \mathbf{r}_{j0}(t) + \mathbf{R}(t)$, where \mathbf{r}_{j0} represents the background thermal motion of each electron, and \mathbf{R} is the superimposed collective motion induced by the optical laser pulse. Without loss of generality, let us assume that we probe the ensemble at the time when all the electrons are at the peak of the laser acceleration (we denote this time as $t = 0$) and hence, for small times $\mathbf{R}(t) = \mathbf{a}t^2/2$, which can be substituted into Eq. (8). In the case of uniform acceleration, during which the motion of the electrons is assumed to be non-relativistic, and ignoring a constant normalization factor, we readily obtain (details provided in the supplementary information)

$$S(\mathbf{k}, \omega) = \frac{c}{\sqrt{\pi\lambda a}} \int S^0(\mathbf{q}, \omega') \exp\left[-\frac{c^2(\omega - \omega')}{\lambda a^2}\right] d\omega', \quad (9)$$

where $S^0(\mathbf{q}, \omega)$ is the usual equilibrium structure factor calculated at $t = 0$, with $\mathbf{q} = \text{Re}(\mathbf{k})$. We notice that for $a \rightarrow 0$ we have $S(\mathbf{k}, \omega) = S^0(\mathbf{q}, \omega)$ as expected.

The case of a dilute electron gas. In order to estimate the relative importance of the acceleration correction to the structure factor, let us consider the case of a weakly interacting electron gas in thermal equilibrium at the temperature T . In this case $S^0(\mathbf{q}, \omega)$ takes a simple Gaussian profile²¹, and from Eq. (9) we obtain

$$S(\mathbf{k}, \omega) = \sqrt{\frac{m}{2\pi q^2 k_B T_{\text{eff}}(q, a)}} \exp\left[-\frac{m}{2q^2 k_B T_{\text{eff}}(q, a)} \left(\omega - \frac{\hbar q^2}{2m}\right)^2\right], \quad (10)$$

where,

$$T_{\text{eff}}(q, a) = T + \lambda \frac{ma^2}{2k_B q^2 c^2}. \quad (11)$$

Eq. (10) also implies that the outgoing photon carries an uncertainty in energy expressed as $\hbar\Delta\omega = \hbar q [k_B(T_{\text{eff}} - T)/m]^{1/2} = \sqrt{2\lambda\pi} k_B T_{DU}$, and is consistent with the energy uncertainty imposed by the motion of the electrons being non-relativistic. Eqs. (7) and (10) are the main result of our work. They show that the scattering of photons from an ensemble of accelerated electrons is seen as if those electrons are in

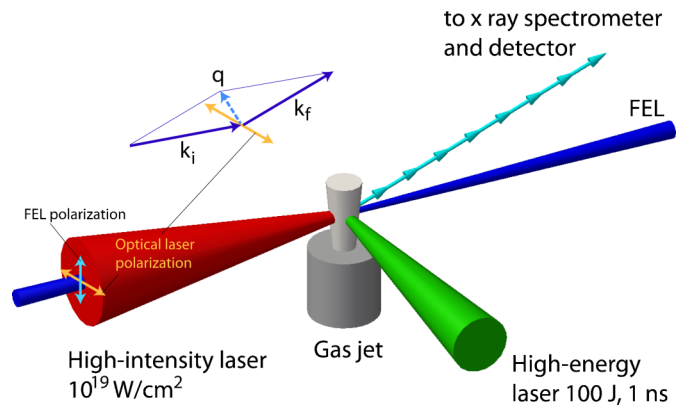


Figure 1 | Proposed experimental setup. A high-energy laser excites a pulsed gas jet creating a plasma with electron density $\sim 10^{19} \text{ cm}^{-3}$. A high-intensity CPA optical laser beam then accelerates the electrons while the FEL probes them with synchronized x ray pulses in a collinear geometry.

thermal equilibrium at an effective temperature T_{eff} , which depends on the acceleration. Moreover, this effective temperature scales with the metric parameter, λ (or ξ), providing, for the first time, a direct way to determine the validity of the models of quantum mechanics in curved space-time, and the specific details of the coupling between classical and quantized fields. Previous work done by solving a generalized form of the Dirac equation in curved space-time has also shown that the metric can have observable effects²². In fact, it was suggested that energy levels shifts in atoms could arise from the local curvature, although at scales not currently achievable in a laboratory.

Discussion

Let us now consider the implication of Eq. (10) in a scattering experiment as laid out in Figure 1. To limit velocity dispersion between the high intensity optical laser and the FEL, the thermal electrons are generated in a $\sim 0.2 \text{ mm}$ long channel by ionization of an underdense low atomic number gas jet (Figure 1), driven by focussing a high energy ($\sim 100 \text{ J}$) long-pulse ($\sim 1 \text{ ns}$) laser beam. The electron densities will be in excess of $\sim 10^{19} \text{ cm}^{-3}$ and temperatures $\sim 200 \text{ eV}$ ²³. We will now assess the feasibility of measuring the effective temperature within current facilities. We write $q^2 \approx 2(\omega_1/c)^2(1 - \mu)$, where μ is the cosine of the scattering angle ($\mu \approx 0.96$ for 15° scattering angle). In practical units,

$$T_{\text{eff}} - T = 1.4 \times 10^{-19} \frac{\lambda I [\text{W/cm}^2]}{(\omega_1 [\text{keV}])^2 (1 - \mu)} \text{ eV} \quad (12)$$

Assuming that the acceleration field is produced with an optical laser with intensity $I \sim 10^{19} \text{ W/cm}^2$, and the thermal electrons (at $T \sim 200 \text{ eV}$) are probed with $\omega_1 \sim 0.5 \text{ keV}$ x ray photons in forward scattering geometry (see Figure 1), we get $T_{\text{eff}} - T \sim 300 \text{ eV}$. This is readily within our experimental capabilities owing to the recent development of Thomson scattering in the x ray regime^{24,25}. Simple estimates indicate that, with $\sim 10^{13}$ photons/pulse in the FEL beam, we should expect > 40 photons/shot detected. A few shots would then be sufficient for obtaining high quality spectra²⁴.

Ideally, we would perform the experiment to probe electrons scattering from the point of maximum acceleration and zero velocity, which sets the pulse duration of the FEL to less than a femtosecond. Recent developments in free electron laser operations have indeed shown that such short pulse duration ($\lesssim 1 \text{ fs}$) in the x ray regime is possible²⁶. At present, the temporal synchronization of optical and x ray pulses has been demonstrated at the few fs level²⁷, but work to reduce it below to $\sim 1 \text{ fs}$ is underway²⁸. Limitation in the pulse duration and temporal synchronization would also introduce a



superimposed chirp in the scattered light due to the varying Doppler shift of electrons at high velocity and modest acceleration. Since the electrons will oscillate primarily along the direction of the optical laser polarization, this effect can be removed in the measured spectrum by choosing the geometry of the experiment such that the scattering wavenumber, q , is perpendicular to the polarization axis, as shown in Figure 1.

While a high power laser, an FEL, as well as a long pulse high-energy laser beam are not currently available on a single end-station, suggestions for such a facility have been already put forward both at the Linac Coherent Light Source (LCLS) in Stanford (CA) and at the European XFEL in Desy (Germany). Indeed, the scientific case described in this letter is very compelling and our estimates indicate that a direct test of the semiclassical theory of quantum mechanics in curved space-time will become possible.

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Author contributions

GG conceived this project. BJBC, with support from GG, performed the calculations and theoretical analysis. BJBC, RB and GG wrote the manuscript. Additional experimental and theoretical contributions were provided by RGE, DOG, OLL, CDM, PAN, SJR, TT, CHTW and JSW.

Additional information

Supplementary information accompanies this paper at <http://www.nature.com/scientificreports>

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Supplementary Information

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Derivation of Equation (7)

The matrix elements of the perturbation operator, $\hat{\mathcal{H}}' \equiv \hat{\mathcal{H}} - \hat{\mathcal{H}}_0 = \mathcal{H}'_{\text{int}}(0)$, defined by equations (4) and (5) in the main paper, between the states (β, i) , denoting the initial state of the system, and (α, f) , denoting a possible final state, in which the labels i, f denote the states of the scattered photon and α, β denote the states of the scatterer system, are straightforwardly given by

$$\begin{aligned} \langle \alpha f | \hat{\mathcal{H}}' | \beta i \rangle &= \frac{e^2}{2mV\epsilon_0} \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \int_V \langle \alpha | \hat{\rho}(\mathbf{r}) e^{-i(\mathbf{k}_f - \mathbf{k}_i - i\boldsymbol{\gamma})\mathbf{r}} | \beta \rangle d^3 \mathbf{r} \\ &= \frac{e^2}{2mV\epsilon_0} \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \langle \alpha | \hat{\rho}_{-\mathbf{q} - i\boldsymbol{\gamma}} | \beta \rangle \end{aligned} \quad (\text{S.1})$$

where $\mathbf{q} = \mathbf{k}_i - \mathbf{k}_f$, $\boldsymbol{\gamma} = 2\mathbf{a}/c^2$ and

$$\hat{\rho}_{\mathbf{k}} \equiv \int_V \hat{\rho}(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} d^3 \mathbf{r} \quad (\text{S.2})$$

Expressing the operators in the interaction picture gives the time evolution of this matrix element according to

$$\begin{aligned}
\langle \alpha f | \hat{\mathcal{H}}'_{\text{int}}(t) | \beta i \rangle &= \frac{e^2}{2mV\epsilon_0} \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \langle \alpha | \hat{\rho}_{-\mathbf{q}-i\boldsymbol{\gamma}}(0) | \beta \rangle \exp(-i(E_{\beta i} - E_{\alpha f})t) \\
&= \frac{e^2}{2mV\epsilon_0} \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \exp(i(\omega_f - \omega_i)t) \langle \alpha | \hat{\rho}_{-\mathbf{q}-i\boldsymbol{\gamma}}(0) | \beta \rangle \exp(-i(\epsilon_i - \epsilon_f)t) \\
&= \frac{e^2}{2mV\epsilon_0} \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \exp(i(\omega_f - \omega_i)t) \langle \alpha | e^{i\hat{\mathcal{H}}_0 t} \hat{\rho}_{-\mathbf{q}-i\boldsymbol{\gamma}}(0) e^{-i\hat{\mathcal{H}}_0 t} | \beta \rangle \\
&= \frac{e^2}{2mV\epsilon_0} \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \exp(i(\omega_f - \omega_i)t) \langle \alpha | \hat{\rho}_{-\mathbf{q}-i\boldsymbol{\gamma}}(t) | \beta \rangle
\end{aligned} \tag{S.3}$$

where $\hat{\mathcal{H}}'_{\text{int}}$ is the perturbation Hamiltonian in the interaction picture, ie as defined by

$\hat{\mathcal{H}}'_{\text{int}}(t) = \exp(i\hat{\mathcal{H}}_0 t) \hat{\mathcal{H}}' \exp(-i\hat{\mathcal{H}}_0 t)$, and $E_{\beta i} = \epsilon_{\beta} + \omega_i$, $E_{\alpha f} = \epsilon_{\alpha} + \omega_f$. Substituting according to (S.3) into equation (6) in the paper, yields

$$\begin{aligned}
\frac{d\sigma_i}{d\Omega_f d\omega_f} &= r_e^2 \frac{4\pi^2 c^3}{V} \mathcal{D}_f \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \sum_{\alpha} \int_{-\infty}^{+\infty} \langle \langle \beta | \hat{\rho}_{\mathbf{q}-i\boldsymbol{\gamma}}(\frac{1}{2}t) | \alpha \rangle \langle \alpha | \hat{\rho}_{-\mathbf{q}-i\boldsymbol{\gamma}}(-\frac{1}{2}t) | \beta \rangle \rangle \exp(i(\omega_i - \omega_f)t) dt \\
&= r_e^2 \frac{4\pi^2 c^3}{V} \mathcal{D}_f \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} \int_{-\infty}^{+\infty} \langle \hat{\rho}_{\mathbf{q}-i\boldsymbol{\gamma}}(-\frac{1}{2}t) \hat{\rho}_{-\mathbf{q}-i\boldsymbol{\gamma}}(\frac{1}{2}t) \rangle \exp(i(\omega_i - \omega_f)t) dt \\
&= \frac{N_e}{V} r_e^2 (2\pi c)^3 \mathcal{D}_f \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} S(\mathbf{q} + i\boldsymbol{\gamma}, \omega) \\
&= \frac{N_e}{V} r_e^2 (2\pi c)^3 \mathcal{D}_f \frac{\mathbf{e}_f \cdot \mathbf{e}_i}{\sqrt{\omega_f \omega_i}} S(\mathbf{k}_i - \mathbf{k}_f + i\boldsymbol{\gamma}, \omega_i - \omega_f)
\end{aligned} \tag{S.4}$$

where $\omega = \omega_i - \omega_f$ and $r_e = e^2/4\pi\epsilon_0 mc^2$ is the classical electron radius and where the Fourier-transformed Van Hove density-density correlation function, aka the dynamic structure factor, is defined by¹

$$\begin{aligned}
S(\mathbf{k}, \omega) &= \frac{1}{2\pi N_e} \int_{-\infty}^{+\infty} \langle \hat{\rho}_{\mathbf{k}^*}(\frac{1}{2}t) \hat{\rho}_{-\mathbf{k}}(-\frac{1}{2}t) \rangle \exp(i\omega t) dt \\
&= \frac{1}{2\pi N_e} \int_{-\infty}^{+\infty} \langle \hat{\rho}_{-\mathbf{k}}^\dagger(\frac{1}{2}t) \hat{\rho}_{-\mathbf{k}}(-\frac{1}{2}t) \rangle \exp(i\omega t) dt
\end{aligned} \tag{S.5}$$

which incorporates the generalisation to *complex* \mathbf{k} . It is readily verified that the function $S(\mathbf{k}, \omega)$ defined by (S.5) is always real for real ω .

Inserting the density of final states, $\mathcal{D}_f = \mathcal{D}(\omega_f) = \frac{V\omega_f^2}{(2\pi c)^3}$, into (S.4) leads to equation (7). Apart from the presence of an imaginary part, γ , this is the known result^{2,3} for the double-differential cross-section for the Thomson scattering of photons by a many-electron system.

We can further generalise the dynamic structure factor (S.5) to time-dependent non-equilibrium situations by means of

$$\begin{aligned}
S(\mathbf{k}, \omega; t) &= \frac{1}{2\pi N_e} \int_{-\infty}^{+\infty} \langle \hat{\rho}_{\mathbf{k}^*}(t + \frac{1}{2}\tau) \hat{\rho}_{-\mathbf{k}}(t - \frac{1}{2}\tau) \rangle \exp(i\omega\tau) d\tau \\
&= \frac{1}{2\pi N_e} \int_{-\infty}^{+\infty} \langle \hat{\rho}_{-\mathbf{k}}^\dagger(t + \frac{1}{2}\tau) \hat{\rho}_{-\mathbf{k}}(t - \frac{1}{2}\tau) \rangle \exp(i\omega\tau) d\tau
\end{aligned} \tag{S.6}$$

in which we have found it convenient to change the integration variable to τ .

Derivation of Equations (9) – (11)

We now consider how the dynamic structure factor (S.6) is modified by the collective acceleration and by \mathbf{k} having an imaginary part. First of all there is the spatial integral involved in Fourier transforming the densities. This leads to a change in the normalisation of the integral that seems to depend on the large-scale spatial inhomogeneity and is thus regarded as indeterminate. We shall not be concerned with this factor.

The time integral is more interesting. To evaluate this, we write the position of the j th particle as

$$\mathbf{r}_j(t) = \mathbf{r}_{0j}(t) + \mathbf{R}(t) \tag{S.7}$$

in which $\mathbf{r}_{0j}(t)$ represents the background thermal motion of each particle and the component $\mathbf{R}(t)$ represents a superimposed collective motion that is independent of the background motion and of the individual particle labels, and where $\mathbf{R}(t)$ is an analytic function of time which is real on $\text{Im}t = 0$.

For a system subject to an initial drift velocity \mathbf{v}_0 and a constant acceleration \mathbf{a} ,

$$\mathbf{R}(t) = \mathbf{v}_0 t + \frac{1}{2} \mathbf{a} t^2 \quad (\text{S.8})$$

Ignoring the effect of large-scale inhomogeneities, we then have

$$\langle \hat{\rho}(\mathbf{r}, t) \hat{\rho}(\mathbf{r}', t') \rangle \simeq \langle \hat{\rho}^0(\mathbf{r} - \mathbf{R}(t), t) \hat{\rho}^0(\mathbf{r}' - \mathbf{R}(t'), t') \rangle \quad (\text{S.9})$$

where $\hat{\rho}^0$ is the ambient equilibrium density, in the absence of any induced collective motion.

By the application of some straightforward algebra, a combination of (S.6), (S.9), (S.2) and (S.8) yields the frequency dependent part of $S(\mathbf{k}, \omega; t)$ according to

$$S(\mathbf{k}, \omega; t) = A(\boldsymbol{\gamma}) \int_{-\infty}^{+\infty} S^0(\mathbf{q}, \omega') F(\mathbf{k}, \omega - \omega'; t) d\omega' \quad (\text{S.10})$$

where $A(\boldsymbol{\gamma})$ is the undetermined normalisation, which is constant (independent of time) for times $t \ll c/a$ provided that $v_0 \ll c$; $S^0(\mathbf{q}, \omega)$ is the equilibrium dynamic structure factor defined by

$$\begin{aligned} S^0(\mathbf{q}, \omega) &= \frac{1}{2\pi N_e} \int_{-\infty}^{+\infty} \langle \hat{\rho}_{-\mathbf{q}}^{0\dagger}(\frac{1}{2}t) \hat{\rho}_{-\mathbf{q}}^0(-\frac{1}{2}t) \rangle \exp(i\omega t) dt \\ &= \frac{1}{2\pi N_e} \int_{-\infty}^{+\infty} \langle \hat{\rho}_{\mathbf{q}}^0(t) \hat{\rho}_{-\mathbf{q}}^0(0) \rangle \exp(i\omega t) dt \end{aligned} \quad (\text{S.11})$$

and

$$\begin{aligned}
F(\mathbf{k}, \omega; t) &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} \exp\left(i\left(\omega\tau - \mathbf{k}^* \cdot \mathbf{R}\left(t + \frac{1}{2}\tau\right) + \mathbf{k} \cdot \mathbf{R}\left(t - \frac{1}{2}\tau\right)\right)\right) d\tau \\
&= \frac{e^{-2\boldsymbol{\gamma} \cdot \mathbf{R}(t)}}{2\pi} \int_{-\infty}^{+\infty} \exp\left(i\left(\omega - \mathbf{q} \cdot \mathbf{v}(t)\right)\tau - \frac{1}{4}\boldsymbol{\gamma} \cdot \mathbf{a} \tau^2\right) d\tau \\
&= \frac{1}{\sqrt{\pi\boldsymbol{\gamma} \cdot \mathbf{a}}} \exp\left(-\frac{1}{\boldsymbol{\gamma} \cdot \mathbf{a}}\left(\omega - \mathbf{q} \cdot \mathbf{v}(t)\right)^2 - 2\boldsymbol{\gamma} \cdot \mathbf{R}(t)\right) \\
&= \frac{1}{\sqrt{\pi\lambda}} \frac{c}{a} \exp\left(-\frac{c^2}{\lambda a^2}\left(\omega - \mathbf{q} \cdot \mathbf{v}(t)\right)^2 - 2\boldsymbol{\gamma} \cdot \mathbf{R}(t)\right) \\
&\simeq \frac{1}{\sqrt{\pi\lambda}} \frac{c}{a} \exp\left(-\frac{c^2}{\lambda a^2}\left(\omega - \mathbf{q} \cdot \mathbf{v}(t)\right)^2\right)
\end{aligned} \tag{S.12}$$

where $\mathbf{k} = \mathbf{q} + i\boldsymbol{\gamma}$, $\lambda = \frac{c^2}{a^2}\boldsymbol{\gamma} \cdot \mathbf{a}$ and $\mathbf{v}(t) = \mathbf{v}_0 + \mathbf{a}t$. Equation (S.12) is in the form of a normalised Gaussian function of ω and tends to the delta function $\delta(\omega - \mathbf{q} \cdot \mathbf{v})$ as $a \rightarrow 0$. Note that convergence of the integration over τ requires that $\lambda \geq 0$, to which all the known metrics conform.

Finally we apply (S.10) in the case of a dilute system represented as a Boltzmann gas of particles of mass m at temperature T , for which the dynamic structure factor is¹

$$S^0(\mathbf{q}, \omega) = \sqrt{\frac{m}{2\pi q^2 T}} \exp\left(-\frac{m}{2q^2 T} \left(\omega - \frac{q^2}{2m}\right)^2\right) \tag{S.13}$$

Performing the convolution (S.10), using (S.12), then yields

$$S(\mathbf{k}, \omega; t) = A(\boldsymbol{\gamma}) \sqrt{\frac{m}{2\pi q^2 T_{\text{eff}}(q)}} \exp\left(-\frac{m}{2q^2 T_{\text{eff}}(q)} \left(\omega - \mathbf{q} \cdot \mathbf{v} - \frac{q^2}{2m}\right)^2\right) \tag{S.14}$$

where

$$T_{\text{eff}}(q) = T + \frac{\lambda m a^2}{2q^2 c^2} \tag{S.15}$$

If the scattering is observed in the plane perpendicular to the laser induced motion, ie the direction of polarisation of the driving laser, then we can set $\mathbf{q} \cdot \mathbf{v} = 0$ in (S.14), which then reduces to the result expressed by equations (9) – (11) in the paper.

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³ M.J. Cooper, P.E. Mijnders, N. Shiotani, N. Sakai and A. Bansil, *X-ray Compton Scattering* (Oxford University Press, 2004)