The speed of information in a ‘fast-light’ optical medium

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One consequence of the special theory of relativity is that no signal can cause an effect outside the source light cone, the space-time surface on which light rays emanate from the source. Violation of this principle of relativistic causality leads to paradoxes, such as that of an effect preceding its cause. Recent experiments on optical pulse propagation in so-called ‘fast-light’ media—which are characterized by a wave group velocity \( v_g \) exceeding the vacuum speed of light \( c \) or taking on negative values—have led to renewed debate about the definition of the information velocity \( v_i \). One view is that \( v_i = v_g \) (ref. 4), which would violate causality, while another is that \( v_i = c \) in all situations, which would preserve causality. Here we find that the time to detect information propagating through a fast-light medium is slightly longer than the time required to detect the same information travelling through a vacuum, even though \( v_g \) in the medium vastly exceeds \( c \). Our observations are therefore consistent with relativistic causality and help to resolve the controversies surrounding superluminal pulse propagation.

The speed of a light pulse travelling through an optical material is not precisely defined, because any pulse comprises a collection of elementary sinusoidal waveforms, each with a distinct frequency \( \omega \). Each constituent sinusoid travels at a well-defined velocity known as the phase velocity \( v_p = c/\eta(\omega) \), where \( \eta(\omega) \) is the refractive index of the optical material. Approximate theories of optical pulse propagation predict that the peak travels at the group velocity \( v_g = c/(\eta + \omega \eta')/\eta(\omega) = c/\eta_g \), where \( \eta_g \) is the group index and \( \omega \) is the central frequency of the wavepacket.

We refer to the quantity \( \eta_g \) as the dispersion of an optical material. For typical optical materials, there exist narrow spectral regions where \( \eta(\omega) \) is a decreasing function of frequency (that is, \( \eta_g < 0 \)), resulting in a condition known as anomalous dispersion. When \( \eta_g \) is within such a region, \( \eta_g \) can be less than one and can even become negative when the anomalous dispersion is large. This results in ‘fast light’, for which it is possible that the peak of a light pulse may exit the optical material before it passes through the entrance face. The amount of fast-light pulse advancement is largest when \( v_g \) is negative and near zero (\( \eta_g \) large and negative).

The possibility of superluminal group velocities (\( v_g > c \) or \( v_g < 0 \)) was such a concern to researchers around 1910 that several conference sessions were devoted to the topic. Based on these discussions, Sommerfeld demonstrated theoretically that the velocity of the front of a square-shaped pulse propagating through any medium is identically equal to \( c \) and hence relativistic causality is preserved. In a follow-up study, Brillouin suggested that the group velocity is not physically meaningful when the dispersion is anomalous because the pulse becomes severely distorted. More recent research investigating the propagation of smooth-shaped pulses has shown that this conclusion is not justified, leading to renewed controversy.

Another outcome of the discussions in the early 1900s, as recounted in the preface and first chapter of the book by Brillouin, was a reformulation of the fundamental postulate of the special theory of relativity. This reformulation states that, rather than limiting the speed of an ‘object’, it is the information velocity \( v_i \) that is limited by \( c \). Unfortunately, there is no agreed-upon definition of the information velocity.
In our experiment, we use a fast-light medium that exploits the spectral region of anomalous dispersion between two closely spaced amplifying resonances\(^{15,17,20}\) realized by creating large atomic coherence\(^{21,22}\) in a laser-driven potassium vapour\(^{23}\), as shown in Fig. 1a. We obtain larger pulse advancement for a smooth gaussian-shaped pulse, as shown in Fig. 1b, in comparison to the experiment of ref. 15, by increasing the gain and hence the size of the anomalous dispersion. The larger advancement relative to the pulse width obtained in our experiment makes it easier to distinguish the different velocities describing pulse propagation. From this data, we infer that \(\eta_0 = -19.6 \pm 0.8\), indicating that we are operating in the highly superluminal regime.

Measuring \(v_i\) requires an understanding of the fundamental mechanism for information encoding and detection. Garrison et al.\(^2\) propose that new information is encoded on an optical pulse by creating a point that is non-analytic (for example, a discontinuity in the pulse amplitude or its derivatives) and that this point always travels at \(c\) regardless of the value of the other velocities associated with the pulse\(^2,5\). Essentially, they have generalized Sommerfeld’s concept of the front velocity to a non-analytic point of the pulse amplitude, where the front of a square-shaped pulse is an example of a point of non-analyticity. These workers\(^{2,5}\) suggest that the point of non-analyticity is the only part of the pulse representing new information because measurements of the early part of the pulse cannot be used to predict anything about the part of the pulse arriving after the point of non-analyticity, and hence \(v_i\) equals the speed of a point of non-analyticity. For counterviews, see refs 2, 4, 23 and 24. We note that some aspects of their proposal have been verified using electronic circuits where no propagating waves are involved, and hence only issues of causality, but not relativistic causality, can be tested\(^{25–27}\).

To enhance our ability to estimate the location of this non-analytic point in the presence of noise, we use two optical pulses that are initially identical gaussian-shaped, which allows us smoothly to turn on the pulse amplitude to a level above the noise floor of our detection electronics and to monitor the fast-light pulse advancement. Near the peak of the gaussian function and at the same moment for both symbols, we switch the amplitude of the gaussian function to a high (1) or low (0) value for the remainder of the pulse. The moment when a decision is made to switch between the symbols corresponds to the point of non-analyticity. Note that this transition is smoothed out by the finite response time of the optical switch.

The location of the point of non-analyticity is determined by detecting the arrival of new information using a receiver that can distinguish between symbols to a desired level of certainty, characterized by the bit error rate (BER). Before the arrival of the point of non-analyticity at the detector, we expect no detected information, corresponding to a BER of 1/2. Once the point of non-analyticity propagates past the detector, the received information will grow smoothly from zero and the BER drops. A symbol is considered to be detected when the BER falls below some threshold. Hence, the detection time of information is later than the time when information is first available at the detector, even for pulses propagating through vacuum. This detection latency \(\Delta t\) depends on the charac-

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**Figure 1** Fast-light pulse propagation. **a**, Experimental set-up. The potassium vapours are contained in two uncoated Pyrex cells of length \(L/2 = 20\) cm to suppress unwanted parametric instabilities\(^{15}\) and heated to obtain an atomic number density of \(4.5 \times 10^{11}\) atoms cm\(^{-3}\). Linearly polarized coherence-preparation laser beams (frequencies \(\omega_{2J}\) and \(\omega_{2J+1}\)) are combined with the linear and orthogonally polarized pulses using polarizing beam splitters. The pulses are detected by a photoreceiver with a 25 kHz–125 MHz bandwidth. The coherence preparation beams are adjusted with the AOM set at 1.36 GHz to the high-frequency side of the centre of the potassium 4S\(^1/2\) — 4P\(^1/2\) transition and \(\omega_{2J} - \omega_{2J+1} = 23\) MHz, chosen to optimize the pulse advancement using procedures similar to those discussed in refs 15, 18 and 22. The pulses are generated by passing a continuous-wave laser beam through an acousto-optic modulator (AOM) driven by a computer-controlled arbitrary waveform generator. The time origin has been set arbitrarily to coincide with the peak of this pulse. **b**, The solid line shows the temporal evolution of a 263.4-ns-long (full-width at half-maximum) pulse propagating through the cells when the lasers are tuned far from the atomic resonance and hence the vapour-cell portion of the path is equivalent to vacuum. The dashed line shows the observed fast-light pulse advancement for a smooth pulse shape when the coherence-preparation laser is tuned near the atomic resonance and \(\omega_{2J}\) is set between the gain resonances. The peak of the pulse is advanced by \(t_{\text{adv}} = 27.4\) ns ± 1.1 ns, corresponding to a relative pulse advancement of 10.4%. Using \(t_{\text{adv}} = Lc/\eta_0\) with \(L/c = 1.3\) ns, we find \(\eta_0 = -0.051 \pm 0.002\). Careful inspection of the fast-light pulse reveals that it has been compressed by 1.9%, which is due primarily to the frequency dependence of the gain\(^{18,19}\).

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**Figure 2** Transmitting information-encoded optical pulses through a fast-light medium. **a**, Transmitting ‘0’ and ‘1’ through the fast-light medium (dashed line) and vacuum (solid line). Each symbol is transmitted separately through the medium and vacuum, where each curve is an average of 50 pulses. **b**, High-resolution plot of part of a with an offset for clarity. The amplitude of the advanced and vacuum pulses have been scaled so that their heights would be the same if a gaussian pulse propagated through the system, as in Fig. 1b. The error bar indicates the typical standard deviation of the pulse amplitudes. From a, it is seen that the fast-light medium advances the early part of the pulses during the smooth turn on, identically to that observed for the full gaussian-shaped pulses shown in Fig. 1b. Most important is the observation that both symbols are the same for early times so that it is not possible to distinguish between them. Hence, no information can yet be conveyed to a receiving party at the end of the communication channel.
To quantify our results, we determine the BER for the vacuum (Fig. 3a, solid line) and advanced (dashed line) pulse pairs using an integrate-and-dump matched filter technique. The BER is high for final observation times in the range between $-40$ and $-25$ ns, during which the pulse amplitudes are large (see Fig. 2a). Hence, even though the signal-to-noise ratio for a single pulse is high at these times, the pulses are not yet distinguishable and no information is detected. Placing the detection threshold at BER $= 0.1$, chosen to keep $\Delta t_{\text{vac}}$ and $\Delta t_{\text{adv}}$ small, we determine the detection time for vacuum (advanced) pulse pairs $T_{\text{vac}}$ ($T_{\text{adv}}$) and the difference in detection times $T_i = T_{\text{adv}} - T_{\text{vac}}$. The time difference is approximately constant for BER values around 0.1; its average value in the range of BERs between 0.08 and 0.2 is equal to $3.2 \pm 1.5$ ns. Our observations demonstrate that the information detection time for pulses propagating through the fast-light medium is longer than the detection time for the same information propagating through vacuum, even though the group velocity is in the highly superluminal regime for the fast-light medium.

From our direct measurement of $T_i$, we do not know whether an observed difference between the detection times is due to changes in the detection latencies or differences in the information velocities for vacuum ($v_{\text{vac}}$) and the fast-light medium ($v_{\text{adv}}$). The relation among these quantities is given by:

$$T_i = \left( I / v_{\text{adv}} - I / v_{\text{vac}} \right) + (\Delta t_{\text{adv}} - \Delta t_{\text{vac}}) \quad \text{(1)}$$

To gain some insight about the importance of detection latency, we analyse a mathematical model, based on Maxwell’s equations, that describes approximately the generation, propagation, and detection of our symbols. Consistent with previous research\textsuperscript{1,3}, this model predicts that $v_{\text{adv}} = v_{\text{vac}} = c$, and hence $T_i$ is completely determined by $(\Delta t_{\text{adv}} - \Delta t_{\text{vac}})$. Using the same matched-filtering approach, we determine the predicted BER as shown in Fig. 3b. We see that information is detected later for the advanced pulses than for the vacuum pulses, qualitatively similar to the experimental observations. We find that $T_i = 1.5 \pm 0.5$ ns, where the error only accounts for statistical uncertainty in the BER determination. The fact that $T_i \neq 0$ demonstrates that subtle changes in the shape of the symbols after information has been encoded give rise to substantial changes in the detection latency. The predicted value of $T_i$ might be smaller than the observed time owing to our assumption that the fast-light medium does not change the noise properties of the optical pulses\textsuperscript{10}.

Using the model prediction for $(\Delta t_{\text{adv}} - \Delta t_{\text{vac}})$ in equation (1) and taking $v_{\text{vac}} = c$, we find that $v_{\text{adv}} = 0.4(\pm 0.7 - 0.2)c$. Thus, our observations are consistent with the special theory of relativity even for a medium where $v_g$ is highly superluminal, demonstrating that the peak of the advanced pulse at the exit face of the medium (see Fig. 1b) is not causally connected to the peak at the entrance face\textsuperscript{2}. Because our analysis makes no assumptions about the sources of noise in the encoding, transmission and decoding process, our general experimental approach and conclusions should hold even in the limit where quantum fluctuations are dominant\textsuperscript{2,29,30}.

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Single-electron transistor of a single organic molecule with access to several redox states

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A combination of classical Coulomb charging, electronic level spacings, spin, and vibrational modes determines the single-electron transfer reactions through nanoscale systems connected to external electrodes by tunnelling barriers. Coulomb charging effects have been shown to dominate such transport in semi-conductor quantum dots, metallic and semiconducting nanoparticles, carbon nanotubes, and single molecules. Recently, transport has been shown to be also influenced by spin—through the Kondo effect—for both nanotubes and single molecules, as well as by vibrational fine structure. Here we describe a single-electron transistor where the electronic levels of a single π-conjugated molecule in several distinct charged states control the transport properties. The molecular electronic levels extracted from the single-electron-transistor measurements are strongly perturbed compared to those of the molecule in solution, leading to a very significant reduction of the gap between the highest occupied molecular orbital and the lowest unoccupied molecular orbital. We suggest, and verify by simple model calculations, that this surprising effect could be caused by image charges generated in the source and drain electrodes resulting in a strong localization of the charges on the molecule.

We measured electrical transport at 4 K through a single p-phenylenevinylene oligomer, which has five benzene rings connected through four double bonds (OPV5, Fig. 1a). OPV5 was placed by chemical vapour deposition on a gap about 2 nm wide separating the source and drain electrodes of a single-electron transistor (SET) device (Fig. 1). The synthesis of OPV5 (EE, 1,4-bis[(E)-(E)-(4,4-(tert-butylthio)styril)]benzene) was done by a Horner–Wadsworth–Emmons condensation of tetraethyl 1,4-xylylenediphosphonate with trans-4-(tert-butylthio)styrilbenzaldehyde. The terminal thiols were protected with a tert-butyl group, which prevents chemical binding of the sulphur to the gold electrode and leads to a weak van der Waals contact between the molecule and the source and drain electrodes.

A planar gate electrode made of aluminium metal covered with aluminium oxide (∼5 nm thick) was prepared on a chip of oxidized silicon. A shadow mask used to deposit the gold lead electrodes was defined on top of the gate by standard electron-beam lithography. The chip was then introduced into a vacuum chamber immersed in liquid helium. All subsequent procedures reported here were performed during a single vacuum cycle. First, two gold electrodes were deposited through a shadow mask by condensing gold vapour on the substrate held at 4.2 K. By using an oblique evaporation angle (Fig. 1b) together with in situ conductance measurements, we were able to fine-tune the tunnelling gap between the gold electrodes to a few nanometres. As shown in Fig. 1b, for an inclined beam, a constriction is formed by two mask edges. If the tilt angle is high, there is no overlap between source and drain shadows. Reducing the tilt angle decreases the source–drain gap. We changed the tilt stepwise, narrowing the gap between the leads by 5 nm at each step. At each step, a 2-nm-thick film of gold was deposited through the mask and the sample was checked for non-zero tunnelling conductance (this procedure of nanometre-scale gap fabrication has been successfully tested for other metals). Eventually, we fabricated two self-aligned and self-sharpened gold electrodes with a tunnelling gap of a few MΩ between them. By annealing the sample up to 100 K, we increased the gap resistance to a few GΩ, which corresponds to a tunnelling gap width of roughly 2 nm (ref. 14). Annealed samples did not show any gate dependence of the tunnelling conductance nor any peculiarities in the current–voltage, I(V), curves. In a separate control experiment, we observed that an irreversible decomposition of gold into separate clusters starts at 150–200 K.

Second, a submonolayer (∼1%) of organic molecules was deposited on the electrodes by quench condensation. The sample was annealed at low temperature (below 70 K) allowing thermally activated motion of the organic molecules, while monitoring the nanogap conductance at a source–drain bias of 400 mV. When the conductance changed stepwise, indicating the trapping of a single molecule in the nanogap, the device was cooled to a temperature of 4.2 K where all transport measurements were taken. This entire process was repeated successfully for three independent devices.

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