Entang-bling: Observing quantum correlations in room-temperature solids.

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Abstract. Quantum entanglement in the motion of macroscopic solid bodies has implications both for quantum technologies and foundational studies of the boundary between the quantum and classical worlds. Entanglement is usually fragile in room-temperature solids, owing to strong interactions both internally and with the noisy environment. We generated motional entanglement between vibrational states of two spatially separated, millimeter-sized diamonds at room temperature. By measuring strong nonclassical correlations between Raman-scattered photons, we showed that the quantum state of the diamonds has positive concurrence with 98% probability. Our results show that entanglement can persist in the classical context of moving macroscopic solids in ambient conditions.

1. Macroscopic quantum correlations

Our intuition about the nature of the physical world is strongly conditioned by the experience that macroscopic bodies move according to the rules of classical mechanics. Quantum theory, however, asserts that superpositions and entanglement are possible even for large objects. Therefore, exploration of the persistence of quantum correlations in the traditionally classical realm is important for both fundamental science and technology, because of the implications for physics beyond conventional quantum mechanics and for quantum information processing, which requires sustained coherence across many particles.

A long-standing challenge is to reveal quantum features in “ordinary” materials under ambient conditions, without a particularly exotic preparation or engineering. The requirements for this are that any experiment must satisfy two conditions. First that it take place on a timescale much shorter than the decoherence time $\tau$ of the degree of freedom in which we expect to observe the correlations. Second that it should take place on a much shorter timescale $\tau' = \tau/n$ than it takes for the degree of freedom to begin to thermalize with its environment, where $n$ is the mean number of excitations in the bath (say a set of bosonic modes) that are coupled to the degree of freedom of interest and cause the decoherence.

Since $\tau$ is small in many solid materials – typical electronic and inter-atomic excitations decays occur on femto- to pico-second timescales – we require a particularly high frequency degree of freedom so that any bath will have a very low thermal occupancy, and thus $\tau' \approx \tau$. Optical phonons provide such a degree of freedom. A number of very tightly bonded crystals have appropriate phonon frequencies and decay times that are accessible using modern ultrafast laser techniques. This allows us both to prepare and to measure single quanta of vibrational excitation.
2. Preparation of nonclassical vibrational states in diamond

Diamond is a particularly promising material for the study of room temperature quantum vibrational states since it has an optical phonon branch with phonon energies up to 1332 cm$^{-1}$, with lifetimes of the order of 10 ps. This combination means that the phonons can be non-impulsively excited with optical pulses of duration great than about 10 fs.

The principle behind the experiment is to use intense, brief, laser pulses to excite the diamond and set it ringing by means of Raman scattering. Raman scattering occurs when the light beam incident on a material is scattered into a different frequency, shifted from that of the incident light by a characteristic frequency of the material. This is typically a vibrational frequency in molecules or crystals. The effect arises from the coupling of the electronic and vibrational degrees of freedom of the material, leading to the dependence of the shift of the electronic resonance frequency of the optical transitions in the material on the amplitude of vibrations. Often Raman scattering is initiated by the thermal population of the material. Crystals, for example, usually have significant population in low frequency vibrational modes at room temperature. However, even at zero degrees Kelvin, when there are no material excitations, Raman scattering can still occur, due to vacuum fluctuations of the electromagnetic field. In that case, a photon from the incident light beam has its energy divided into a single down-shifted photon - the Stokes photon - and a single quantum of excitation of the material. Thus the number of Stokes photons and the number of material quanta are tightly correlated.

We used a modelocked Ti:Sapphire laser, producing 80 fs duration pulses at 805 nm mean wavelength, to excite optical phonons near the center of the Brillouin zone of a diamond crystal. Diamond has a particularly large Raman scattering cross-section, and an electronic band gap of several eV, making it well suited to optical manipulation. During scattering, the diamond absorbs some energy from the laser pulse and begins to vibrate. The photon emerging from the diamond, called the Stokes photon, is red-shifted, with less energy than an incident laser pulse photon. The detection of a Stokes photon signals that an optical phonon has been created in the diamond.

Similarly, the reverse process, whereby a photon from the incident light beam gains energy from the material to scatter into a higher-energy, shorter-wavelength photon - the anti-Stokes photon - may also occur, once the material has some excitation. The ratio of Stokes to anti-Stokes intensity provides therefore a means to measure the degree of excitation of the material. We next send in another laser pulse, and we look for an anti-Stokes photon — that is, a blue-shifted Raman scattered photon. The detection of a Stokes photon signals that an optical phonon has been created in the diamond.

Of course, in practice, the correlations
are never perfect – detector noise, poor mode matching, background light all contribute to the signal. However, it was possible to show that the probability with which anti-Stokes photons were detected once a Stokes photon had been detected was much greater than any classical model would allow.

The outcome of a measurement of the Stokes/anti-Stokes correlations are shown in Fig. 1 [1]. The strength of the correlations is specified by the ratio $g^{(2)}_{S,\alpha S}$ of the joint probability $P_{S,\alpha S}$ for detecting both Stokes and anti-Stokes photons, to the product of the marginal probabilities for detection of either Stokes photons ($P_S$) or anti-Stokes photons ($P_{\alpha S}$):

$$g^{(2)}_{S,\alpha S} = \frac{P_{S,\alpha S}}{P_S P_{\alpha S}}.$$ 

Classically, the magnitude of the correlation is bounded by the Cauchy-Schwartz inequality $g^{(2)}_{S,\alpha S} \leq \sqrt{g^{(2)}_{S,S} g^{(2)}_{\alpha S,\alpha S}}$, where the autocorrelations of the Stokes and the anti-Stokes fields appear under the radical. These fields are approximately thermal, with $g^{(2)}_{S,S} = g^{(2)}_{\alpha S,\alpha S} = 2$, so that measured values of $g^{(2)}_{S,\alpha S}$ exceeding 2 are indicative of nonclassicality. This allows us to infer that the vibrational state generated by the scattering of a Stokes photon is itself nonclassical. Indeed, the results are consistent with it being a single vibrational quantum, as shown in Fig. 1.

3. Entangling the vibrational states of two diamonds

This approach can be extended to prepare a single phonon distributed across two diamonds. This “sharing” of a single quantum of excitation between the two spatially separated crystals is described by an entangled state.

This may be understood by an optical analog, and shown in Fig. 2(a). A single photon incident on a beam splitter may exit by either port. Since there is only a single photon, and the mode transformation is unitary, it ends up in a superposition: it has a probability amplitude for being found in either mode, with a definite phase relation between the two amplitudes. The combination of properties of a single excitation of the optical field (just one photon) distributed between the two paths (in a coherent superposition state of the two modes) is a sufficient condition to prove entanglement [2].

This may be confirmed experimentally by reversing the process, illustrated in Fig. 2(b). If a single photon in a path superposition state is incident on the beam splitter, then the photon will always exit a particular port. Which port is determined by the phase difference between the two paths. This can be adjusted by inserting a phase shifter in one of the paths, so that the photon always exits a specific port. A photon that occupied either path at random (described by a mixed state) would exit through the two output ports at random, in an uncontrollable fashion.

Of course revealing path interference by adjusting phases between the two paths is not in itself a specifically quantum interference – two coherent states occupying the paths would exhibit the same phenomenon. But, when combined with a
measurement of $g^{(2)}_{\text{SS}}$ of the conditionally prepared anti-Stokes beam, this is enough to show entanglement, provided the photon statistics are sub-Poissonian. The density operator of the state in the photon number basis then shows ideally nothing in the zero and two photon sectors ($p_{00} = p_{22} = 0$), but full quantum coherence in the one photon sector ($p_{10}, p_{01} \neq 0$). This means that the concurrence of the state, defined as $C = 2 \max \left(0, \sqrt{V(p_{10} + p_{01})/2} - \sqrt{p_{00}p_{22}}\right)$, is positive.

The same measurement strategy may be used to demonstrate the entanglement of two diamonds by means of a shared phonon. The arrangement is shown schematically in Fig. 3. First, a single phonon is generated in a pair of diamonds by illuminating them with two laser pulses generated by splitting a single input pulse on a beamsplitter. The spontaneous scattering of a single Stokes photon from the pair of incident laser pulses heralds the excitation of a single vibrational quantum. Erasing the which-path information of the Stokes photon by means of a beam splitter leaves that the diamond in which the phonon resides unknowable, even in principle. Thus the quantum state describing the diamonds is a superposition state in which the phonon is in one diamond and the other at the same time. The Stokes photon may be detected at either output port, and this varies randomly from shot to shot.

The state of the phonon in the two diamonds can be determined by “reading it out” using anti-Stokes Raman scattering. The single phonon is converted into a single photon by this means. If the anti-Stokes photon always exits the beamsplitter by the same port as the Stokes photon did, then this is sufficient proof that the diamonds were entangled, because it shows that the anti-Stokes photon was in an entangled state.

Of course, experimental imperfections mean that this is never the case, but a measurement of the concurrence of the state of the anti-Stokes beam by the methods described above is possible, and shows that the anti-Stokes photon is entangled. Fig. 4 shows the experimentally measured density matrix of the anti-Stokes beam, conditioned on the detection of a single Stokes photon. The diagonal elements are direct measurements of the zero-, one- and two-photon probabilities (shown in blue, red and green, respectively). The one-photon sector off-diagonal elements are obtained from the visibility of the interference fringes obtained by adjusting the relative phase of the two anti-Stokes paths by means of a waveplate. The grey elements were unmeasured components of the density matrix. These are difficult to access for small photon number states (they require, for instance, an operation that can rotate a two-photon state into a one-photon state, for
instance). However, for our purposes we can set them to zero and still obtain a lower bound on the degree to which the anti-Stokes beam is in an entangled state. If they are non-zero, the entanglement only increases [2]. Since we have already shown that the anti-Stokes photons are tightly correlated with the existence of a phonon in the diamonds, we can infer that the diamonds are entangled [3].

3.1. Collapse vs retrodiction

One feature of our experiment that can be puzzling is the fact that the time for the Stokes and anti-Stokes photons to propagate to the detector and thus to be registered is much longer than the lifetime of the phonons in the diamonds, which have decohered after less than 10 picoseconds.

In quantum physics, dynamics and measurements are generally decomposed into a set of temporally instantaneous operations (unitaries and projectors) acting globally on Hilbert space without explicit reference to space-time. Such analysis can lead to the seemingly straightforward notion that a measurement on part of a distributed system (say, the registration of a photon emitted from one of the diamonds at point D in Fig. 5) causes an instantaneous collapse of the global quantum state across all spatial positions at the same moment (point C in Fig. 5). However the idea that the registration of a Stokes photon “prepares” the single phonon state in the diamond by “collapsing” the wavefunction is problematic [3]. A causal effect would “generate” the entanglement at a later time within the light cone of point A – say point F in Fig. 5.

The answer to this enigma is well understood, but is nonetheless worth rehearsing since the untenable position of action at a distance implied by the collapse hypothesis is rather starkly illustrated by our apparatus. The collapse interpretation leads to some uncomfortable conclusions that are strongly at odds with a standard notion of causality. Specifically, in other inertial reference frames, such spatially separated “collapse” events would occur at different times. Indeed, it is always possible to find reference frames in which the quantum state at other locations collapses before the detection event itself takes place. In fact, measurements in quantum
mechanics are not related to causality; projection operations update the state to enforce consistency with classical data. In general, a measurement projection can always be back-propagated to any arbitrary time during the system evolution [4]. In the case of photodetection, the physically significant time is the moment at which the photons were created, and the purpose of such a measurement is to provide information about the system at this instant. In our experiment, it is therefore natural to treat the propagation of signals by evolving the Stokes measurement operator backwards in time from the moment of detection, \( \tau_0 \), (point D) to the time \( \tau_S \) (point A) at which the Stokes photon was scattered. The resulting projection leaves the diamonds in an entangled state at \( \tau_S \), and it is this entanglement that is probed by anti-Stokes scattering at time \( \tau_A = \tau_S + T \), where \( T \) is less than the decoherence time of the phonon.

4. Conclusions
We have experimentally demonstrated that two room-temperature solids can be prepared in an entangled state. In order to show this we combined ultrafast interferometric pump-probe scheme with photon-counting techniques to generate and prepare short-lived quantum correlations in a strongly decohering environment. This approach lays the foundation for future studies of quantum phenomena in many-body, strongly interacting systems in ambient conditions, which may prove to be helpful in understanding the emergence of the classical world from the quantum.

Acknowledgements
This work was undertaken at the University of Oxford with support from EPSRC, the Royal Society and the European Union. B. J. Sussman and D. England are currently at the National Research Council of Canada in Ottawa, K. C. Lee is at TeraView, Cambridge, and N. Langford is at Royal Holloway College, London.

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