Superluminal and Slow Light Propagation in a Room-Temperature Solid

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states by mapping onto correlated nonclassical states of light, raising the possibility of applications in quantum information science.

Note added in proof: We have been made aware of related work recently published by Kuzmich *et al*. (*39*).

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34. Experimental details are available as supporting material on *Science* Online.
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37. In the case where optical fields have losses before detection, the level of twin-mode squeezing that can be observed is reduced (*32*). For our data, the anti-Stokes signal appears identical to the Stokes signal, except for an apparent attenuation by a factor of ~5 and a time shift (Fig. 2C). In this case, twin-mode squeezing can still be investigated when the time shift is compensated and when the unbalanced attenuation is compensated by unbalanced linear amplification of the two signals. We have developed a method based on fast, high-accuracy sampling of the Raman signals followed by software compensation of the time shift and linear amplification. Accordingly, this procedure accounts for the rescaling of photon shot-noise and detector dark-noise levels. The resulting signals are subtracted for evaluation of the spectrum in Fig. 4. For further details, see (*40*).
38. The criterion for two modes of light to be photon number squeezed is that for a sequence of photon number measurements, n_1 and n_2, the variance in $(n_1 - n_2)$ must be less than the sum of the averages $(\bar{n}_1 + \bar{n}_2)$ (*32*). For measurements on two continuous photon flux signals, analyzed in the frequency domain, this corresponds to the fluctuation spectrum of the time-domain difference signal being less than such a fluctuation spectrum from two classical fields with the same average photon flux and classical intensity correlations, such as would be obtained by splitting a single light beam on a perfect beam splitter.
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41. Experimental work was carried out at the Harvard University Department of Physics. We thank T. P. Zibrova, J. MacArthur, M. Hohensee, S. B. Shenai, P. R. Hemmer, and A. Trifonov for useful discussions and experimental help. Supported by NSF grant PHY-0113844, the Defense Advanced Research Projects Agency, the David and Lucille Packard Foundation, the Alfred Sloan Foundation, the Office of Naval Research (DURIP program), the Smithsonian Institution, the MIT-Harvard Center for Ultracold Atoms, a fellowship through the Netherlands Organization for Scientific Research (C.H.v.d.W.), and a NSF Graduate Research Fellowship (M.D.E.).

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Superluminal and Slow Light Propagation in a Room-Temperature Solid

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We have observed both superluminal and ultraslow light propagation in an alexandrite crystal at room temperature. Group velocities as slow as 91 meters per second to as fast as −800 meters per second were measured and attributed to the influence of coherent population oscillations involving chromium ions in either mirror or inversion sites within the crystal lattice. Namely, ions in mirror sites are inversely saturable and cause superluminal light propagation, whereas ions in inversion sites experience conventional saturable absorption and produce slow light. This technique for producing large group indices is considerably easier than the existing methods to implement and is therefore suitable for diverse applications.

Propagation of light pulses through material systems has been studied quantitatively for almost a century (*1*). It has been understood that the group velocity (v_g) of a pulse passing through a resonant system could be slower than the velocity of light in a vacuum (c) or could even be faster than c without contradicting the causality principle (*2*–*5*). Recently, techniques have been developed that have brought the control over the group velocity of light to such extremes as ultraslow light ($v_g << c$) (*6*–*9*), fast light ($v_g > c$ or v_g is negative) (*10*–*13*), and "stored" or "stopped" light (*14*, *15*). These techniques hold promise for uncovering new physical phenomena and for practical applications such as controllable optical delay lines, optical data storage, optical memories, and devices for quantum information.

In the first experimental observation of slow and fast light propagation in a resonant system (*16*), laser pulses propagated without appreciable shape distortion but experienced very strong resonant absorption (~10^5 cm^{-1}). To reduce absorption, most of the recent work on slow light propagation has used electromagnetically induced transparency (EIT) to render the material medium highly transparent (*17*, *18*) and still retain the strong dispersion required for the creation of slow light. Using this technique, a group velocity of $v_g = c/165$ in a 10-cm-long Pb vapor cell was observed (*6*). More recently, a group velocity of 17 m/s was found for propagation in a Bose-Einstein condensate (*7*), and a modulation technique was used to measure a group velocity of 90 m/s in Rb vapor (*8*). Using similar techniques, group velocities as low as 8 m/s have been inferred (*9*). Solid-state materials have also been used to observe the propagation of slow light with a velocity of 45 m/s through Pr-doped Y_2SiO_5, maintained at a cryogenic temperature of 5 K (*19*).

There also has been considerable work in the production of fast light. Electromagnetically induced absorption has been used

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to observe a group velocity of $-c/23{,}000$ (*11*), while gain-assisted superluminal light propagation (*12*) has been used to demonstrate a group velocity of $-c/310$ (*13*). However, all of these techniques for producing both fast and slow light require complicated experimental configurations and/or low temperatures.

In contrast to these techniques, we recently used a different quantum effect—coherent population oscillations—to produce a group velocity of 58 m/s at room temperature in ruby (*20*). Although our absorption ($\sim$1 cm^{-1}) was higher than the absorption in the EIT experiments, it was several orders of magnitude lower than that in the earlier resonant systems (*16*).

We report an observation of both ultraslow ($v_g << c$) and superluminal ($v_g < 0$) light propagation at room temperature in an alexandrite crystal. By using a modulation technique, we can vary the delay seen by a modulated beam from 440 to –50 μs. We explain these results as the combination of different absorption cross sections and lifetimes for Cr^{3+} ions at either mirror or inversion sites within the $BeAl_2O_4$ crystal lattice. The superluminal wave propagation is produced by a narrow "antihole" [612 Hz half width at half maximum (HWHM)] in the absorption spectrum of Cr^{3+} ions at the mirror sites of the alexandrite crystal lattice, and the slow light originates from an even narrower hole (8.4 Hz) in the absorption spectrum of Cr^{3+} ions at the inversion sites.

To develop an understanding of our experiment, we will first clarify how the optical properties of a material are modified by coherent population oscillations. The first experimental investigation (*21*) of spectral holes caused by coherent population oscillations observed an extremely narrow spectral hole with a width of 37 Hz (HWHM). An Ar-ion laser operating at 514.5 nm was used to pump population from the ground state to the broad $^4F_2$ absorption band of ruby. Electrons decay from this band to a metastable level within a few picoseconds, and eventually return to the ground level with a population relaxation time (T_1) of a few milliseconds. A second laser beam, the probe beam (which may take the form of amplitude modulation sidebands of the pump beam), causes the electron population to oscillate between the ground and metastable levels at the beat frequency δ between the pump and probe beams. However, because the relaxation time is long, these oscillations occur with appreciable amplitude only if the beat frequency δ is so small that $\delta T_1 \sim 1$. When this condition is met, the pump wave can efficiently scatter off the temporally modulated ground state population into the probe wave, resulting in reduced absorption of the probe wave. The shape of the probe absorption profile is given by (*21*)

$$\alpha(\delta) = \frac{\alpha_0}{1+I_0}\left[1 - \frac{I_0(1+I_0)}{(T_1\delta)^2 + (1+I_0)^2}\right] \quad (1)$$

where α_0 is the unsaturated absorption coefficient and I_0 is the pump intensity normalized to the saturation intensity. Because the probe beam experiences decreased absorption over a very narrow frequency interval (of a width on the order of $1/T_1$), the refractive index increases rapidly over this same frequency region. This change in refractive index associated with any absorption feature is a well-known optical phenomenon and is described formally by the Kramers-Kronig relations (*22*). As a result of the rapid spectral variation of $n(\delta)$, the group index $n_g = n_0 + \omega dn(\delta)/d\delta$ is also very large. Here, n_0 is the refractive index of the probe (1.74 for alexandrite) and ω is the optical frequency. Correspondingly, the group velocity $v_g = c/n_g$ is very small. This is the procedure that was implemented to produce slow light propagation using the extremely narrow spectral feature in ruby (*20*).

Whereas a spectral hole leads to slow light, an antihole, that is, a narrow spectral region of increased absorption, leads to fast light. The reason is that, again as a consequence of the Kramers-Kronig relations, the refractive index decreases rapidly with frequency in the region of an antihole, leading to a negative value of the group index. Such an antihole, with a measured width of 612 Hz, has been observed in alexandrite at 457 nm (*23*). An antihole is formed rather than a hole because in the range 450 to 510 nm, alexandrite displays inverse-saturable absorption resulting from the strong excited-state absorption (*24*). However, an antihole is not the only spectral feature within that wavelength range; a hole can be seen as well. Alexandrite is formed by doping a $BeAl_2O_4$ crystal with Cr^{3+} ions, and these ions replace the Al^{3+} ions (*25*), but not all of the ion sites are identical. Namely, 78% of the sites occupied by the Cr^{3+} ions have mirror symmetry (C_s), and the rest have inversion symmetry (C_i) (*26*). As a result, the absorption cross sections σ_1, population relaxation times T_1, and saturation intensities $I_s \equiv \hbar\omega/\sigma_1 T_1$ are different for the two different sites (Fig. 1). Ions at mirror sites have a relaxation time of 290 μs, and ions at inversion sites have a relaxation time of ~50 ms (*26*). Our measurements suggest that the mirror-site ions have a large excited-state absorption cross section σ_2, whereas the inversion-site ions experience negligible excited-state absorption. We reached this conclusion because the width of the antihole (which is a consequence of excited-state absorption) is the inverse of 290 μs, whereas the width of the spectral hole (which does not involve excited-state absorption) is approximately the inverse of 50 ms.

In our experiment (fig. S1), we use an Ar-ion laser that we operate at either 476 or 488 nm. The beam passes first through a variable attenuator and then an electro-optic modulator. The modulator is driven by a function generator that places a sinusoidal amplitude modulation on the beam. The side modes created by the modulation act as probe beams that interact with the spectral features created by the pump. A glass slide sends 5% of the beam to one detector for reference. The beam is then focused with a 20-cm–focal length lens near the front surface of a 4.0-cm-long alexandrite crystal. The orientation of the crystal is such that the light is polarized parallel to the *a* axis. The transmitted beam passes through an interference filter to remove any fluorescence from

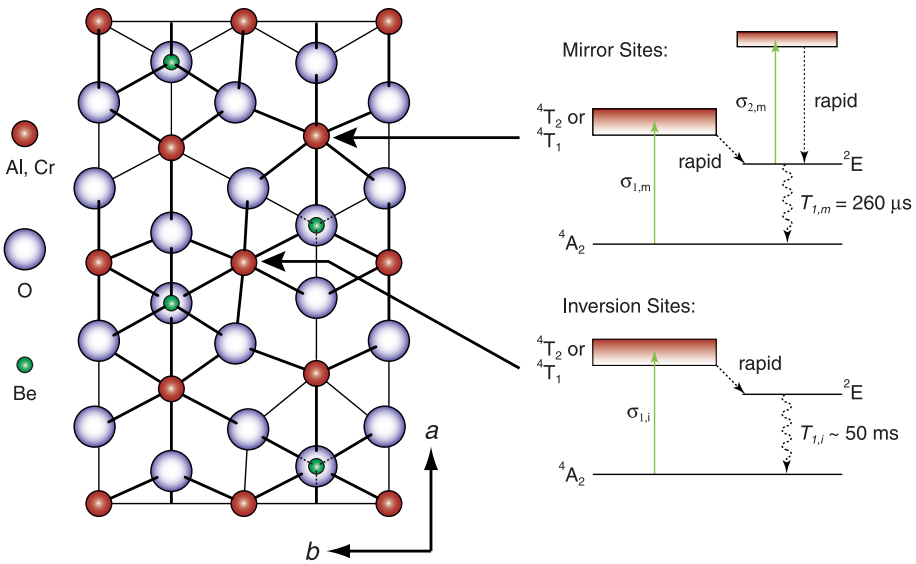

Fig. 1. The crystal structure of alexandrite looking along the *c* axis (*25*). The arrows indicate the locations of ion sites that have mirror or inversion symmetry. On the right, the corresponding energy-level diagrams for Cr^{3+} ions at the different sites are shown. Mirror-site ions experience excited-state absorption and have a population relaxation time of 260 μs. Inversion-site ions have negligible excited-state absorption and a much longer population relaxation time (~50 ms).

electrons decaying from level ^2E to the ground state and then falls onto a detector. The detected signal is stored along with that of the reference beam on a digital oscilloscope, and the resulting traces are compared to determine the relative delay and amplitude of the two signals.

To model our results, we consider the influence of ions both at the inversion sites and at the mirror sites. In addition, the absorption cross sections are assumed to be different at different wavelengths. With these considerations, we calculate numerically the intensity throughout the crystal and use this result to find the expected delay and attenuation of the modulated signal. We find the best fit for our data using the ion number density $N = 9 \times 10^{19}$ cm^{-3} and population relaxation times $T_{1,m} = 280$ μs, $T_{1,i} = 19$ ms, where the m and i subscripts indicate the parameter for a mirror or inversion site. We give the absorption cross sections that we used for each wavelength below. All of these parameters are within the range of accepted values (*24*, *26*).

As we have described above, the slope of the probe absorption profile ($d\alpha/d\delta$) determines the group velocity: A positive slope tends to produce slow light, a negative slope fast light. We independently measured the probe absorption and modulation delay as functions of frequency, and we display both of them to demonstrate the self-consistency of our experimental data. In Fig. 2, we show both the relative modulation attenuation, defined as the attenuation of the modulated signal (probe) relative to the attenuation of the constant background (pump), and the delay for pump powers of 250 and 410 mW at a wavelength of 476 nm. We observe an advancement of the waveform as large as 50 μs, which corresponds to a group velocity of −800 m/s and a group index of -3.75×10^5. The relative modulation attenuation (Fig. 2A) that we measure is similar to what is observed in (*23*) at 457 nm; however, the influence of inversion-site ions can be seen in the small dip in the absorption at low modulation frequencies. The measured transmission was about 3.5%. In our modeling of these results, we used the values $\sigma_{1,i} = 0.35 \times 10^{-20}$ cm^2, $\sigma_{1,m} = 0.9 \times 10^{-20}$ cm^2, and $\sigma_{2,m} = 4.05 \times 10^{-20}$ cm^2.

Finally, in Fig. 3 we show the results obtained at a wavelength of 488 nm. At this wavelength, the effect of the inversion sites dominates and produces a very narrow dip in the absorption at low frequencies. The peak in the delay, shown in Fig. 3B, corresponds to an average group velocity of 148 m/s, but we also observed group velocities as low as 91 m/s with a higher pump power (950 mW). The transmission at this wavelength was more than 10%. As can be seen from the inset in Fig. 3B, we get good agreement with our numerical model at this wavelength using the parameters $\sigma_{1,i} = 0.4 \times 10^{-20}$ cm^2, $\sigma_{1,m} = 0.9 \times 10^{-20}$ cm^2, and $\sigma_{2,m} = 3.5 \times 10^{-20}$ cm^2.

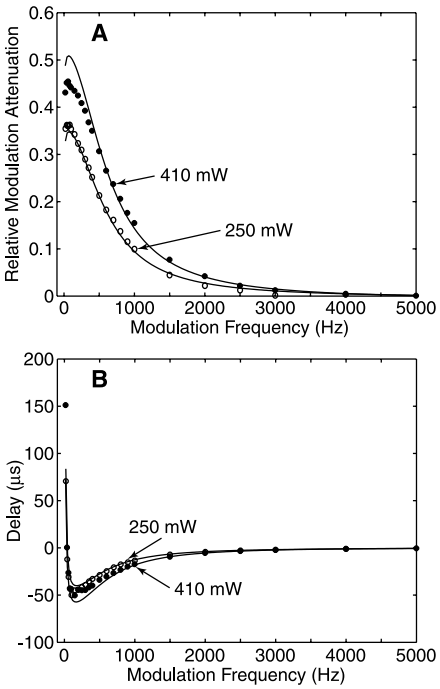

Fig. 2. (**A**) Relative modulation attenuation and (**B**) time delay measured for a 4-cm-long alexandrite crystal at a wavelength of 476 nm with pump powers of 250 and 410 mW. The observed negative time delay corresponds to superluminal propagation. The solid lines indicate the results of our theoretical model.

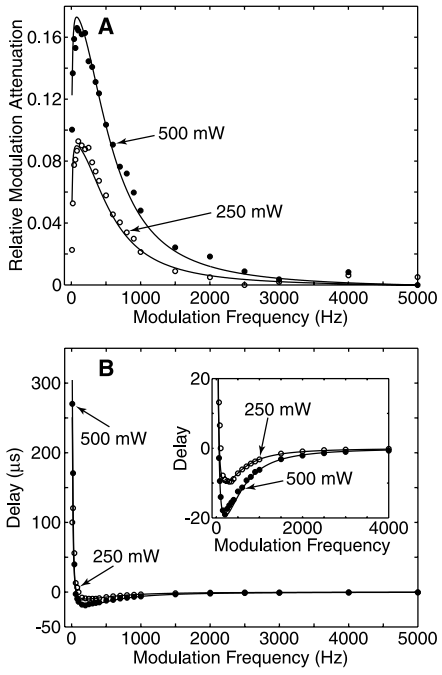

Fig. 3. (**A**) Relative modulation attenuation and (**B**) time delay measured for a 4-cm-long alexandrite crystal at a wavelength of 488 nm with pump powers of 250 and 500 mW. Ultraslow propagation occurs for low modulation frequencies (<60 Hz) and superluminal propagation occurs at higher frequencies. The inset in (B) is a close-up of the same data. The solid lines indicate the results of our theoretical model.

We have demonstrated that either ultraslow or superluminal light propagation can be achieved in the same solid-state material by changing the excitation wavelength. This phenomenon occurs as a result of coherent population oscillations between the ground and excited states in an alexandrite crystal. We find that we have to take account of the different absorption characteristics of Cr^{3+} ions in mirror or inversion sites to interpret our results. In this report, we have demonstrated a delay or advancement of a modulated signal. However, as we displayed in ruby (*20*), this concept can be extended to pulses.

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Fig. S1

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