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24. The SOHO/MDI project is supported by NASA grant NAG5-10483 to Stanford University. SOHO is a project of international cooperation between ESA and NASA. L.R. was supported as a summer intern at NASA/Marshall Space Flight Center through the Marshall Space Grant Research Internship Project.

15 September 2009; accepted 13 January 2010
10.1126/science.1181990

Cavity Quantum Electrodynamics with Anderson-Localized Modes

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A major challenge in quantum optics and quantum information technology is to enhance the interaction between single photons and single quantum emitters. This requires highly engineered optical cavities that are inherently sensitive to fabrication imperfections. We have demonstrated a fundamentally different approach in which disorder is used as a resource rather than a nuisance. We generated strongly confined Anderson-localized cavity modes by deliberately adding disorder to photonic crystal waveguides. The emission rate of a semiconductor quantum dot embedded in the waveguide was enhanced by a factor of 15 on resonance with the Anderson-localized mode, and 94% of the emitted single photons coupled to the mode. Disordered photonic media thus provide an efficient platform for quantum electrodynamics, offering an approach to inherently disorder-robust quantum information devices.

The interaction between a single photon and a single quantized emitter is the core of cavity quantum electrodynamics (QED) and constitutes a node in a quantum information network (*1*, *2*). So far, cavity QED experiments have been realized with a wide range of two-level systems, including atoms (*3*), ions (*4*), Cooper-pair boxes (*5*), and semiconductor quantum dots (*6*–*8*) coupled to photons confined in a cavity. A common requirement for all these implementations is highly engineered cavities, in some cases requiring nanometer-scale accuracy (*9*). Surprisingly, multiple scattering of photons in disordered dielectric structures offers an alternative route to light confinement. If the scattering is very pronounced, Anderson-localized modes form spontaneously. Anderson localization (*10*) is a multiple-scattering wave phenomenon that has been observed for, e.g., light (*11*), acoustic waves (*12*), and atomic Bose-Einstein condensates (*13*). We have demonstrated cavity QED with Anderson-localized modes by efficiently coupling a single quantum dot (QD) to a disorder-induced cavity mode (*14*) in a photonic crystal waveguide.

Photonic crystals are composite nanostructures in which a periodic modulation of the refractive index forms a photonic band gap of frequencies where light propagation is fully suppressed. By deliberately introducing a missing row of holes in a two-dimensional photonic crystal membrane, the periodicity is broken locally and light is guided (Fig. 1A). Such photonic crystal waveguides are strongly dispersive, i.e., light propagation depends sensitively on the optical frequency and can be slowed down. Engineering the photonic crystal waveguide enables the enhancement of light-matter interaction, which is required for high-efficiency single-photon sources (*15*) for quantum information technology (*1*, *16*). In the slow-light regime of photonic crystal waveguides, light propagation is very sensitive to unavoidable structural imperfections (*17*, *18*) and multiple-scattering events randomize propagation (*19*). Although multiple scattering is commonly considered a nuisance for a device, leading to optical losses, here the influence of wave interference in multiple scattering stops light propagation and forms strongly confined Anderson-localized modes (*10*) (Fig. 1B). Anderson-localized modes in a photonic crystal waveguide appear as a result of the primarily one-dimensional nature of the propagation of light provided that the localization length is shorter than the length of the waveguide (*20*).

We deliberately created Anderson-localized modes by fabricating photonic crystal waveguides with a lithographically controlled amount of disorder (Fig. 1B). The hole positions in three rows above and below the waveguide were randomly perturbed with a standard deviation varying between 0 and 6% of the lattice parameter. We investigated the Anderson-localized modes by recording QD photoluminescence spectra under high-excitation power where the feeding from multiple QDs makes Anderson-localized modes appear as sharp spectral resonances (Fig. 1C). The observation of spectrally separated random resonances is a signature of Anderson localization of light (*14*), while the detailed statistics of the intensity fluctuations unambiguously verifies localization even in the presence of absorption (*21*). Figure 1D shows the intensity distribution from spectra recorded at different spatial and spectral positions, which allow us to average over different realizations of disorder. Clear deviations from the Rayleigh distribution predicted for nonlocalized waves are observed. Light is localized if the variance of the normalized intensity fluctuations exceeds the critical value of 7/3 (*21*), and we extract a variance of 5.3, which proves Anderson localization.

Examples of Anderson-localized modes are shown in Fig. 2 as peaks appearing at random spectral positions, although limited to the slow-light regime of the photonic crystal waveguide. The latter property is due to the strongly dispersive behavior of the localization length that is considerably shortened in the slow-light regime. We tuned the spectral range of Anderson-localized modes by controlling the amount of disorder. Even in samples without engineered disorder, intrinsic and thus unavoidable imperfections, such as surface roughness, are sufficient to localize light (*22*).

The important cavity figures-of-merit are the mode volume *V* and the *Q* factor. Decreasing *V* leads to an enhancement of the electromagnetic field and thus improves light-matter coupling. The *Q* factor is proportional to the cavity storage time of a photon that needs to be increased for cavity QED applications. High *Q* factors ranging between 3000 and 10,000 are obtained for different degrees of disorder (Fig. 2) and are comparable to state-of-the-art values obtained for traditional photonic crystal nanocavities containing QDs (*7*).

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31. We gratefully acknowledge T. Schlereth and S. Höfling for quantum dot growth, J.M. Hvam for discussions, and the Council for Independent Research (Technology and Production Sciences and Natural Sciences) and the Villum Kann Rasmussen Foundation for financial support.

Supporting Online Material
www.sciencemag.org/cgi/content/full/327/5971/1352/DC1
Materials and Methods
References

24 November 2009; accepted 20 January 2010
10.1126/science.1185080

Light-Controlled Self-Assembly of Semiconductor Nanoparticles into Twisted Ribbons

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The collective properties of nanoparticles manifest in their ability to self-organize into complex microscale structures. Slow oxidation of tellurium ions in cadmium telluride (CdTe) nanoparticles results in the assembly of 1- to 4-micrometer-long flat ribbons made of several layers of individual cadmium sulfide (CdS)/CdTe nanocrystals. Twisting of the ribbons with an equal distribution of left and right helices was induced by illumination with visible light. The pitch lengths (250 to 1500 nanometers) varied with illumination dose, and the twisting was associated with the relief of mechanical shear stress in assembled ribbons caused by photooxidation of CdS. Unusual shapes of multiparticle assemblies, such as ellipsoidal clouds, dog-bone agglomerates, and ribbon bunches, were observed as intermediate stages. Computer simulations revealed that the balance between attraction and electrostatic repulsion determines the resulting geometry and dimensionality of the nanoparticle assemblies.

Spirals, helicoids, helices, twisted ribbons (TRs), and other helical structures present fascinating geometries from the perspectives of mathematics, biology, optics, and mechanics. The formation of helices from nanoparticles (NPs) will make possible exploitation of the unusual properties of helices arising from quantum confinement (*1*, *2*) within NPs, as well as expand the design space (*3*, *4*) and offer new means of controlling the pitch and/or chirality of the helical structures. To achieve self-organization of such intricate objects, it is necessary to fine-tune the overall balance of forces, including anisotropy of interactions that drive assembly of NPs into larger structures. Control of these processes will require the discovery of many-body interactions at the nanoscale, as well as understanding their dynamics and capabilities of formation of complex self-organized patterns transitioning from nano- to microscale.

As a model system for realizing these goals, we used an aqueous dispersion of CdTe (*5*) NPs (emission maximum at 550 nm) prepared (*6*) with thioglycolic acid (TGA) as a stabilizer with the TGA-to-Cd^{2+} ratio close to 1.0, rather than the traditional value of 2.4 (*7*). Based on the variety of existing data (*1*, *6*, *8*), these NPs exhibit strong anisotropy due to permanent dipoles on them (*1*, *8*). The strongly reduced concentration of TGA is expected to lead to the elimination of tetrahedral apices, where the local concentration of TGA is the highest, and an increase of the average value of dipole on NPs. It also increases chemical reactivity of the NPs, which intricately interplays with the interparticle forces. After preparation, the CdTe NPs are precipitated by addition of methanol and centrifuged for 20 min, followed by redispersion in deionized water at pH = 9 (adjusted by addition of NaOH). The orange color of the NP solution turns dark green within ~72 hours, indicating that NP self-assembly has occurred.

TRs with distinctive helicity (Fig. 1, A, B, and D to F), were the primary product of the aging process (Fig. 1) (*6*). The length of the TRs made from CdTe typically ranged from 0.8 to 2 μm, but can be as long as 8 μm (Fig. 1). Some straight nanowires (NWs) were also produced as a secondary product (Fig. 1C) and were identified as well-studied single crystalline Te wires (fig. S1) (*7*). Unlike these NWs or other NP assemblies (*6*), the TRs were made from individual NPs layered on top of each other (Fig. 1, G to I). Their thickness, as determined by atomic force microscopy (AFM), was 10 to 12 nm and corresponds to three to four NP layers. The pitch of the CdTe TRs averaged ~350 nm (Fig. 1, E and F). The distribution between right- and left-handed twisting was approximately equal: 52% right and 48% left, which indicates a nearly racemic mixture of chiral isomers (fig. S2) (*7*). Remarkably, the helical ribbons form bundles in which all TRs have the same chirality. Instead of the typical red shift of optical features found in previous studies of NP assemblies (*6*, *9*, *10*), a gradual blue shift of the luminescence and absorption peaks was observed during formation of TRs (Fig. 2A). This observation normally would have indicated the decrease in delocalization volume of excitons in CdTe and, hence, a wholly different process is taking place than that previously reported. This phenomenon is rather unusual but can be understood when the entire process of transformation of NPs to helicoidal structures is discussed.

We first characterized the composition of the products with x-ray energy dispersive spectroscopy (XEDS). The atomic percent Cd:Te:S ratio for TRs and original CdTe NPs was 46:10:44 and 43:38:19, respectively. The assembly process is associated with considerable loss of Te and transition to CdS/CdTe NPs, in which the CdS phase is strongly dominant. This substantial change of

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