Coupling nanocrystals to a high-Q silica microsphere: Entanglement in quantum dots via photon exchange

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Coupling nanocrystals (quantum dots) to a high-Q whispering gallery mode (WGM) of a silica microsphere, can produce a strong coherent interaction between the WGM and the electronic states of the dots. Shifting the resonance frequencies of the dots, for instance by placing the entire system in an electric potential, then allows this interaction to be controlled, permitting entangling interactions between different dots in a way analogous to the ion-trap computer of Cirac and Zoller. Thus, a more advanced system of this type could potentially be used to implement a simple quantum computer.

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Recent years have seen a dramatic improvement in the ability to experimentally manipulate and measure single quantum systems, from modes of the electromagnetic field to atoms or ions in traps to increasingly tiny solid-state devices. These efforts have been further spurred by developments in the field of quantum computation, where it has been shown that a computer making full use of quantum mechanics can solve certain problems (such as factoring and database searching) faster than any known classical algorithm. Building such a computer requires well-isolated but highly controllable quantum systems, with well-defined levels that can serve as quantum bits (qubits).

A number of proposals have been made for physical implementations of a quantum computer. Two of the most promising involve using the internal electronic states of ions [1] or atoms [2] in a trap as qubits, with single-qubit operations performed by laser pulses; interactions between qubits would involve an intermediate ''bus,'' which would be the quantized motion in the case of ions, an electromagnetic mode of a high-Q cavity in the case of atoms. In a similar spirit, Sherwin *et al.* [3] recently proposed using semiconductor quantum dots (QDs) as qubits, with interactions mediated by the field of an optical microcavity. An earlier proposal by Loss and DiVincenzo [4] also involved QDs, in that case using the spin of a single electron in a QD, with coupling between neighboring dots due to the exchange interaction. Recently, a hybrid of these two proposals has also been suggested by Imamoglu *et al.* [5], in which interactions between electron spins of different dots are mediated by one mode of a microcavity.

In this paper we propose and analyze a novel system for deterministic generation of quantum entanglement of two QDs, using a variant of this same approach. In this system, semiconductor nanocrystals are attached to the equator of a fused-silica microsphere. Exchange of photons between the two nanocrystals through optical interactions between a nanocrystal and a high-Q whispering gallery mode (WGM) of the microsphere leads to creation of quantum entanglement between the two nanocrystals. Entanglement of nanocrystals with slightly different resonance frequencies can be generated by using an electric field to tune the relevant transition frequency of a given nanocrystal to resonate with the WGM. The ground state and the lowest, dark excited state of the nanocrystals serve as a qubit, and it is these levels that become entangled. While the current system is unsuitable for general quantum computations, due to relatively high dephasing rates and the optical frequency difference between energy levels of the dots, with carefully tailored QDs that may be available in the near future, the proposed scheme has the potential to scale up to implement at least a small quantum computer.

Fused-silica microspheres are perhaps the best available optical microresonators [6]. In these microspheres, WGMs form via total internal reflection along the curved boundary. Slight deformation of typical microspheres also removes azimuthal degeneracy of the WGMs. Lowest-order WGMs are thus a ring along the equator of the microsphere. For a fused-silica sphere with a diameter of 20 μm, the very small mode volume leads to a vacuum electric field (or field strength per photon) of order 150 V/cm at the sphere surface ($\lambda \approx 600$ nm). Q factors of these microresonators can exceed 10^9, corresponding to a photon storage lifetime near a microsecond and resulting in highest finesse for optical resonators. Extremely high Q factors along with very small mode volume are essential for achieving strong coherent light-matter coupling, and have made fused-silica microspheres an attractive alternative to the high finesse Fabry-Perot microresonators currently used in cavity QED studies of atoms.

Recent experimental attempts to take advantage of WGMs in fused-silica microspheres to achieve strong coherent light-matter coupling have included putting a fused-silica microsphere in atomic vapors [7] or placing a semiconductor nanostructure in the evanescent waves of WGMs of a microsphere [8]. Further development, however, has been hindered by thermal motion of the atoms in the former case and by

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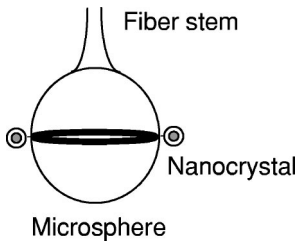

FIG. 1. Schematics of a combined nanocrystal-microsphere system where CdS-capped CdSe nanocrystals are attached to the surface of a fused-silica microsphere.

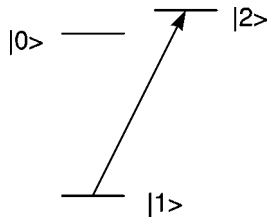

FIG. 2. Schematics of the energy level structure used. $|1\rangle$, the ground state, and $|0\rangle$, the lowest excited state and a dark state, serve as a qubit. The $|1\rangle$ to $|2\rangle$ transition is dipole allowed. Higher excitations are neglected.

significant Q spoiling occurring at the contact area in the latter case. To avoid these problems, we propose to directly attach semiconductor nanocrystals to the equator of a fused-silica microsphere, as shown in Fig. 1.

Semiconductor nanocrystals exhibit discrete atomiclike excitonic energy structures [9]. Recent advances in colloidal chemistry have led to synthesis of epitaxially grown, nearly defect free, CdSe/CdS core/shell nanocrystals and synthesis of high quality III-V nanocrystals including GaAs, InAs, and InP nanocrystals [10]. Near unity quantum yield has been observed in epitaxially grown CdSe nanocrystals even at room temperature. Photoluminescence studies of single CdSe nanocrystals have revealed extremely narrow linewidth, limited by instrument resolution [11]. Assuming a radiative lifetime of 10 ns for a dipole transition in CdSe nanocrystals, we expect the coherent coupling rate between the dipole transition and a resonant WGM with a vacuum field of 150 V/cm to be of order 10^9/sec, much greater than the radiative decoherence rate for the relevant transition.

In semiconductor nanocrystals, the precise excitonic level spacing depends sensitively on the exact size and shape of the nanocrystals. In general two different nanocrystals will have different resonance frequencies although these resonance frequencies can be precisely determined. This nonuniformity of nanocrystals, however, can be turned into an advantage. If one could shift the resonance frequencies of all the nanocrystals simultaneously, it would be possible to bring a single nanocrystal into resonance with a cavity mode while the others are all sufficiently detuned to have no significant interaction with the cavity mode.

One way of producing such a shift is to put the entire system between two microelectrodes and apply a controlled voltage. The level spacings would all be shifted by the quantum confined Stark effect of nanocrystals [12]. Provided that the responses of the various nanocrystals involved have been determined by measurement, it should be possible to tune the system to bring any nanocrystal into resonance on demand. If the voltage changes are slow compared to optical frequencies, this resonance shift may be treated adiabatically. (The voltage change may still be very rapid compared with the timescale of interactions between the internal state of the nanocrystals and the cavity mode.)

It is also possible to drive transitions between the internal levels of individual nanocrystals by applying laser pulses tuned to their particular resonance frequencies, or possibly by using the applied voltage to bring the dots into resonance with fixed lasers. (Since these frequencies will be distinct, tightly focusing the beams onto single dots should be unnecessary.) By combining such laser pulses with carefully timed interactions with the cavity mode, one can construct a scheme very similar to that of Cirac and Zoller [1] or Pellizzari *et al.* [2], in which laser pulses act as one-qubit ''gates,'' while two-bit gates use the photon mode as an intermediary ''bus.'' Exploiting the frequency differences between dots may greatly simplify near-term experiments with small numbers of dots.

We now consider two nanocrystals interacting with a nearly resonant high-Q WGM. We model the two QDs as three-level systems as shown in Fig. 2, where states $|0\rangle$ and $|1\rangle$ serve as the logical states of the qubit and $|2\rangle$ is an auxiliary state. State $|0\rangle$ is the lowest excited state and a dark state [13]. Other energy levels are far off resonance for the relevant optical interactions and are thus neglected. The resonance frequency of the $1\rightarrow 2$ transition is assumed to be close but not equal to the frequency of the cavity mode. This resonance frequency is assumed to differ for the two dots by an amount $\Delta\omega$. The entire system is then placed between two microelectrodes, so that a precisely controlled voltage can be applied, shifting the frequencies of the two nanocrystals.

The two dots are each coupled to the WGM with a strength g. It is assumed that the time scale $1/g$ is long compared to the optical time scales. We change the voltage slowly compared to optical time scales, but quickly compared to $1/g$; thus, this change can be treated adiabatically as far as the Hamiltonian is concerned, but as instantaneous compared to the rate at which photons are emitted or absorbed.

A product state for this model can then be written

$$|\Psi\rangle = |\phi_A\rangle \otimes |\phi_B\rangle \otimes |\phi_\gamma\rangle, \tag{1}$$

where $|\phi_A\rangle$ is the state of dot A, $|\phi_B\rangle$ is the state of dot B, and $|\phi_\gamma\rangle$ is the state of the cavity mode. In a rotating frame we can then write the Hamiltonian of this system

$$\begin{aligned}\hat{H} = & ig(|2\rangle\langle 1| \otimes \hat{1} \otimes \hat{a} - |1\rangle\langle 2| \otimes \hat{1} \otimes \hat{a}^\dagger) \\ & + ig(\hat{1} \otimes |2\rangle\langle 1| \otimes \hat{a} - \hat{1} \otimes |1\rangle\langle 2| \otimes \hat{a}^\dagger) \\ & + \delta(t)\hat{1} \otimes |1\rangle\langle 1| \otimes \hat{1} \\ & + [\delta(t) - \Delta\omega]|1\rangle\langle 1| \otimes \hat{1} \otimes \hat{1}. \end{aligned} \tag{2}$$

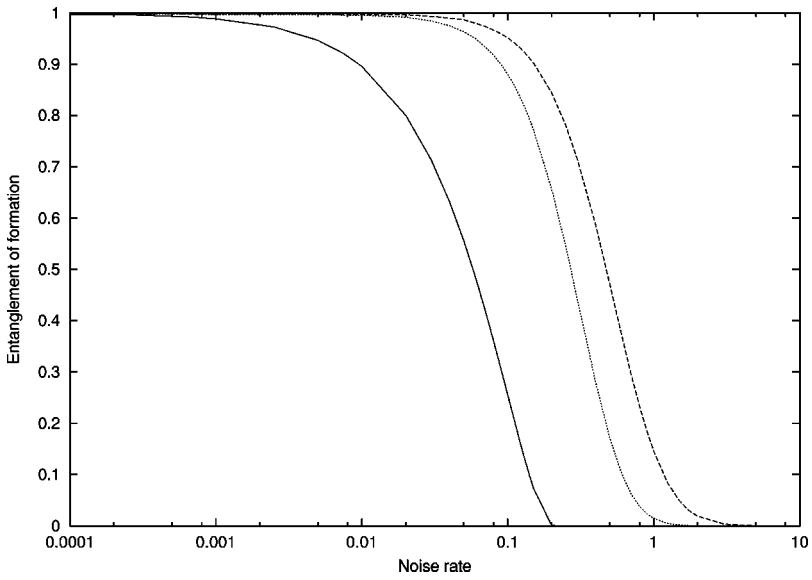

FIG. 3. Entanglement of formation vs noise rate for decoherence between $|0\rangle$ and $|1\rangle$ (solid line), radiative decay from $|2\rangle$ to $|1\rangle$ (dashed line), and cavity loss of the WGM (dotted line). The noise rate is given as the fraction Γ/g, where g is the dipole coupling strength between the nanocrystal and the cavity mode.

The function $\delta(t)$ is the detuning produced in the $1\rightarrow 2$ resonance due to the time-varying voltage. Either A or B (or neither) can be brought into resonance with the cavity mode, but not both at once. We assume that their frequency difference $\Delta\omega$ is large compared to g.

We begin in a product state

$$|\Psi_i\rangle=\frac{1}{2}(|0\rangle+|1\rangle)\otimes(|0\rangle+|1\rangle)\otimes|0\rangle \tag{3}$$

and apply a π pulse to the $1\rightarrow 2$ transition of dot A. (The coherent superposition state of each dot can be created by using resonant two-photon absorption.) The voltage is tuned such that $\delta(t)=\Delta\omega$ for a time $\pi/2g$, then $\delta(t)=0$ for a time π/g, then back to $\delta(t)=\Delta\omega$ for another $\pi/2g$. Finally, a second π pulse is applied to dot A. In the absence of noise, the final state becomes

$$|\Psi_f\rangle=\frac{1}{2}(|0\rangle\otimes|0\rangle+|0\rangle\otimes|1\rangle+|1\rangle\otimes|0\rangle-|1\rangle\otimes|1\rangle)\otimes|0\rangle. \tag{4}$$

From a product state, the two dots have evolved to a state of maximal entanglement. The full set of transformations are summarized here,

$$\begin{aligned}
&|0\rangle|0\rangle|0\rangle\rightarrow|0\rangle|0\rangle|0\rangle\rightarrow|0\rangle|0\rangle|0\rangle\rightarrow|0\rangle|0\rangle|0\rangle,\\
&|1\rangle|0\rangle|0\rangle\rightarrow-i|1\rangle|0\rangle|1\rangle\rightarrow-i|1\rangle|0\rangle|1\rangle\rightarrow|1\rangle|0\rangle|0\rangle,\\
&|0\rangle|1\rangle|0\rangle\rightarrow|0\rangle|1\rangle|0\rangle\rightarrow|0\rangle|1\rangle|0\rangle\rightarrow|0\rangle|1\rangle|0\rangle,\\
&|1\rangle|1\rangle|0\rangle\rightarrow-i|1\rangle|1\rangle|1\rangle\rightarrow i|1\rangle|1\rangle|1\rangle\rightarrow-|1\rangle|1\rangle|0\rangle.
\end{aligned} \tag{5}$$

In practice, noise or decoherence places severe limits on the entanglement process discussed above. We can model the effects of noise by replacing Schrödinger's equation with a Markovian master equation for the system,

$$\frac{d\rho}{dt}=-i[\hat{H},\rho]+\sum_k 2\hat{L}_k\rho\hat{L}_k^\dagger-\hat{L}_k^\dagger\hat{L}_k\rho-\rho\hat{L}_k^\dagger\hat{L}_k, \tag{6}$$

where $\hat{H}$ is the Hamiltonian (2) above, and the $\hat{L}_k$ are a set of operators chosen to model the effects of the noise. We consider three different cases: decoherence between $|0\rangle$ and $|1\rangle$, where

$$\hat{L}_1=\sqrt{\Gamma}|0\rangle\langle 0|\otimes\hat{1}\otimes\hat{1}, \tag{7}$$

$$\hat{L}_2=\sqrt{\Gamma}\hat{1}\otimes|0\rangle\langle 0|\otimes\hat{1}, \tag{8}$$

radiative decay between $|1\rangle$ and $|2\rangle$, where

$$\hat{L}_1=\sqrt{\Gamma}|1\rangle\langle 2|\otimes\hat{1}\otimes\hat{1}, \tag{9}$$

$$\hat{L}_2=\sqrt{\Gamma}\hat{1}\otimes|1\rangle\langle 2|\otimes\hat{1}, \tag{10}$$

and cavity loss, where

$$\hat{L}_1=\sqrt{\Gamma}\hat{1}\otimes\hat{1}\otimes\hat{a}. \tag{11}$$

Γ represents the noise rate. The important parameter in assessing the effects of noise is the size of Γ compared with g; if Γ is small, we expect entanglement to still be possible. (Of course, in general, all three types of noise might well be present, with different Γs; we are modeling the case where one form of noise dominates over the others.)

Figure 3 plots the entanglement of formation [14] as a function of Γ/g for the three different types of noise. With $\Gamma\ll g$, substantial entanglement between the two dots can still be retained. As shown in Fig. 3, the entanglement is the least tolerant to decoherence between $|0\rangle$ and $|1\rangle$ and is the most tolerant to decay from $|1\rangle$ to $|2\rangle$, since the latter decoherence occurs only for the relatively short duration of the relevant optical transitions. Since $|0\rangle$, the lowest excited state, is also a dark state, a ratio $\Gamma/g<10^{-3}$ can be achieved for decoherence between $|0\rangle$ and $|1\rangle$ if the decoherence is

limited by radiative decay. For a Q factor near 10^9, a ratio Γ/g near 10^{-3} is also expected.

To probe whether a nanocrystal is in the ground or the dark excited state, one can, for example, shift the nanocrystal resonance near to a given WGM resonance and measure the induced change in the resonant frequency of the WGM. The WGM resonance remains unchanged when the nanocrystal is in a dark excited state since the dipole transition is bleached. Significant change in the resonant frequency of the WGM results when the nanocrystal remains in the ground state. This gives a readout only of the individual qubit states, which does not allow one to detect entanglement directly. However, by examining the statistics of measurement results as we perform rotations on the internal states of the qubits, it should be possible to establish coherence; this is analogous to the procedure used in experiments on entanglement in ion traps [15].

More ambitious quantum computations are in principle possible, but a number of difficulties would have to be overcome. For the QDs we have discussed, the excitonic levels are separated by optical frequencies, producing a rapidly rotating relative phase between $|0\rangle$ and $|1\rangle$. Very precise timing would be required to control for this, making the experiments prohibitively difficult. The simplest solution would be to use QDs with two closely spaced levels to serve as $|0\rangle$ and $|1\rangle$; this may be possible with doped nanocrystals in the near future.

If this problem can be solved, the above scheme could be scaled up for quantum information processing. One could select and attach N nanocrystals, each with different but known resonance frequencies, to the equator of a single microsphere. Laser pulses could be tuned to any of the dots individually, while an applied voltage could bring any of the dots into resonance with the WGM. Quantum logic gates such as the one described above could be applied between any two dots by exchange of a photon, while laser pulses would enable a large family of rotations of the internal states of the QDs. Together, these two kinds of gates are sufficient to perform quantum computation. The length of such computations will be limited by inevitable decoherence processes, of course.

The experimental challenge of implementing the proposed scheme is still considerable. The above theoretical analysis has not included decoherence in QDs due to electron-phonon interactions. While in ideal QDs with strong three-dimensional confinement and for the lowest excited states, population relaxation due to electron-phonon interactions can be suppressed by the discrete density of states of the system [16], electron-phonon interactions can still lead to rapid decoherence through a pure dephasing mechanism [17,18]. In this regard, III-V nanocrystals are preferable since these nanocrystals can feature weaker electron-phonon interactions than II-VI nanocrystals. Nanocrystals with larger oscillator strength and hence greater coherent dipole coupling rates are also preferable. By operating at very low temperatures, small Γ/g ratios are still achievable [19]. Other obstacles are surface related problems, including charge or field fluctuations on the surface of nanocrystals [13] and relaxation of optical excitations into or via surface states. With continued progress toward fabricating ideal nanocrystals especially in epitaxial capping of nanocrystals, a microcavity system with nanocrystals or artificial atoms covalently bounded to a fused-silica microsphere should eventually provide a realizable model system for studies of quantum entanglement of QDs and for implementing at least a modest number of quantum logic gates. Note that preliminary studies have already demonstrated an extremely high Q factor for combined nanocrystal-microsphere systems [20]. It is possible that suspended nanocrystals may have much lower pure dephasing rates than embedded QDs, due to quantization in phonon energy. Further experimental studies are needed to assess this and other possibilities.

Finally, we note that the combined nanocrystal-microsphere system discussed above not only takes advantage of the long-lived lowest excited state in a QD as well as one of the best available optical microresonators, but also allows separate fabrication and assembly of individual QDs and microspheres. This unique flexibility of ''quantum assembly'' can be an important advantage over epitaxially-grown quantum-dot-microcavity systems.

In conclusion, we have demonstrated theoretically one possible scheme for producing entanglement, and suggest that this type of physical system is a promising candidate for quantum information processing. It may well be possible to adapt other procedures to this system, such as the recent proposal of Imamoglu *et al.* that takes advantage of very long spin decoherence time [5]. Experiments should bring answers to the questions that remain.

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