

sample can increase its local order and lower its net energy by subdividing into finite clusters of strongly interacting particles, with relatively weak exchange of thermal properties between clusters. Furthermore, the sample can increase its entropy by forming aggregates of statistically indistinguishable clusters, with an average number of clusters per aggregate of  $\bar{n} = \partial \ln(\sum_{n=1}^{\infty} \Gamma^n/n!)/\partial \ln \Gamma$  (ref. 5). Evidence for anomalous clustering can be found in measurements of magnetic hysteresis<sup>16</sup>, angle-resolved photoemission<sup>17</sup> and neutron scattering<sup>18</sup>, but perhaps the most direct demonstration of thermodynamic heterogeneity comes from the technique of non-resonant spectral hole burning, NSHB<sup>19</sup>. Observation of NSHB requires the energy absorbed in a selected degree of freedom to remain localized for at least as long as its slow response, which is interpreted as selective local heating. Magnetic NSHB has been observed in a spin glass<sup>20</sup> and in single crystals of EuS and Fe (ref. 21).

The solid symbols in Fig. 1 show the measured magnetic correlation range ( $\xi$ ) in crystalline Co (ref. 22). Assuming aggregates of compact clusters, a theoretical expression for  $\xi$  varies as  $(\bar{m}\bar{n})^{1/3}$ . Indeed,  $(\bar{m}\bar{n})^{1/3}$  exhibits a temperature dependence that is similar to  $\xi$ , but its amplitude is about a factor of ten too large. At least part of this discrepancy comes from the inadequacy of the Ising model for real spins. When normalized to give one atomic radius as  $T \rightarrow \infty$  (solid grey curve in Fig. 1), there is quantitative agreement between  $(\bar{m}\bar{n})^{1/3}$  and  $\xi$  with no new parameters. Thus I have shown that the failure of classical mean-field theories to yield local correlations, and the failure of homogeneous models to yield the measured critical exponents, can be resolved by applying essentially the same mean-field theory to finite clusters with unrestricted sizes. □

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# Universal quantum computation with the exchange interaction

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Various physical implementations of quantum computers are being investigated, although the requirements<sup>1</sup> that must be met to make such devices a reality in the laboratory at present involve capabilities well beyond the state of the art. Recent solid-state approaches have used quantum dots<sup>2</sup>, donor-atom nuclear spins<sup>3</sup> or electron spins<sup>4</sup>; in these architectures, the basic two-qubit quantum gate is generated by a tunable exchange interaction between spins (a Heisenberg interaction), whereas the one-qubit gates require control over a local magnetic field. Compared to the Heisenberg operation, the one-qubit operations are significantly slower, requiring substantially greater materials and device complexity—potentially contributing to a detrimental increase in the decoherence rate. Here we introduced an explicit scheme in which the Heisenberg interaction alone suffices to implement exactly any quantum computer circuit. This capability comes at a price of a factor of three in additional qubits, and about a factor of ten in additional two-qubit operations. Even at this cost, the ability to eliminate the complexity of one-qubit operations should accelerate progress towards solid-state implementations of quantum computation<sup>1</sup>.

The Heisenberg interaction has many features<sup>2,5</sup> that have led to its being chosen as the fundamental two-qubit interaction in a large number of recent proposals for the implementation of quantum computation. Its functional form is very accurate—deviations from the isotropic form of the interaction, arising only from relativistic corrections, can be very small in suitably chosen systems. It is a strong interaction, so that it should permit very fast gate operation, well into the GHz range for several of the proposals. At the same time, it is very short-ranged, arising from the spatial overlap of electronic wavefunctions, so that it should be possible to have an on–off ratio of many orders of magnitude. Unfortunately, the Heisenberg interaction by itself is not a universal gate<sup>6</sup>, in the sense that it cannot generate any arbitrary unitary transformation on a collection of spin-1/2 qubits. So, every proposal has supplemented the Heisenberg interaction with some other means of

applying independent one-qubit gates (which can be thought of as time-dependent local magnetic fields). But the need to add this capability to the device adds considerably to the complexity of the structures, by putting unprecedented demands on “g-factor” engineering of heterostructure materials<sup>4,7</sup>—requiring that strong, inhomogeneous magnetic fields be applied<sup>2,5</sup>—or involving microwave manipulations of the spins that may be slow and may cause heating of the device<sup>4</sup>. These added complexities may well exact a high cost, perhaps degrading the quantum coherence and clock rate of these devices by orders of magnitude.

The reason that the Heisenberg interaction alone does not give a universal quantum gate is that it has too much symmetry: it commutes with the operators  $\hat{S}^2$  and  $\hat{S}_z$  (for the total spin angular momentum and its projection on the  $z$  axis), and therefore it can only rotate among states with the same  $S$ ,  $S_z$  quantum numbers. But by defining coded qubit states—ones for which the spin quantum numbers always remain the same—the Heisenberg interaction alone is universal<sup>8–10</sup>, and single-spin operations with all their attendant difficulties can be avoided.

Recent work has identified the coding required to accomplish this. Early work identified techniques for suppressing phase-loss mechanisms due to coupling with the environment<sup>11–13</sup>, and more recent studies have identified encodings that are completely immune from general collective decoherence—in which a single environmental degree of freedom couples in the same way to all the spins in a block. These codes are referred to both as decoherence-free subspaces (and their generalization, the decoherence-free subsystems)<sup>8,10,14</sup>, and also as noiseless subspaces and subsystems<sup>9,15,16</sup>. The noiseless properties of these codes are not relevant to the present work; but they have the desired property that they consist of states with definite angular-momentum quantum numbers.

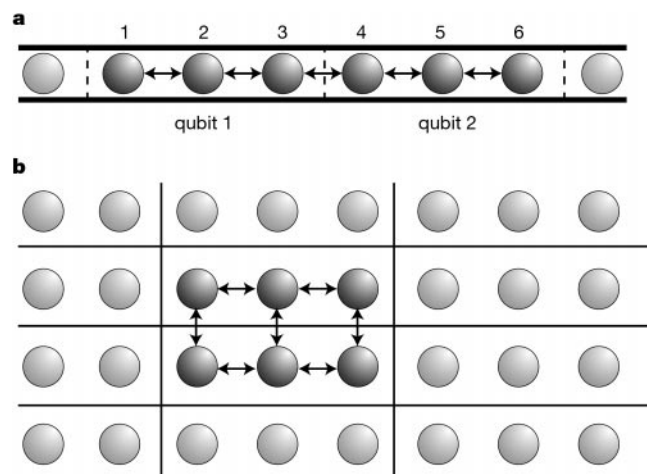
So, in principle, the problem has been solved: the Heisenberg interaction alone is universal and can be used for quantum computation. However, a very practical question still remains: how great is the price that must be paid in return for eliminating single-spin operations? In particular, how many applications of the Heisenberg interaction are needed to complete some desired quan-

tum gate operation? The only guidance provided by the existing theory<sup>8–10</sup> comes from a theorem of Solovay and Kitaev (refs 17, 18, and R. Solovay, personal communication), which states that “efficient” approximations exist: given a desired accuracy parameter  $\epsilon$ , the number  $N$  of exchange operations required goes like  $N \approx K \log^c(1/\epsilon)$ , where  $c \approx 4$  and  $K$  is an unknown positive constant. However, this theorem provides very little useful practical guidance for experiment; it does not show how to obtain the desired approximating sequence of exchange operations, and, as  $K$  is unknown, it gives no clue to whether the number of operations needed for a practical accuracy parameter is 10 or 10,000. Here we remedy these inadequacies by showing that the desired quantum logic operations can be obtained exactly, using sequences of exchange interactions short enough to be of practical significance for forthcoming experiments.

In the scheme we analyse here, we use the smallest subspace with definite angular-momentum quantum numbers that can be used to encode a qubit; this subspace is made up of three spins. We note<sup>10</sup> that in principle the overhead in spatial resources could be made arbitrarily small: the rate of encoding into such noiseless subsystems converges asymptotically to unity. The space of three-spin states with spin quantum numbers  $S = 1/2$ ,  $S_z = +1/2$  is two-dimensional, and will serve to represent our coded qubit. A good explicit choice for the basis states of this qubit are  $|0\rangle = |S\rangle|\uparrow\rangle$ ,  $|1\rangle = (2/3)^{1/2}|T_+\rangle|\downarrow\rangle - (1/3)^{1/2}|T_0\rangle|\uparrow\rangle$ . Here  $|S\rangle = (1/2)^{1/2}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$  is the singlet state of spins 1 and 2 (see Fig. 1a) of the three-spin block, and  $|T_+\rangle = |\uparrow\uparrow\rangle$  and  $|T_0\rangle = (1/2)^{1/2}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$  are triplet states of these two spins. For these states we have constructed an explicit exchange implementation of the basic circuit elements of quantum logic<sup>6</sup>; in particular, we discuss how to obtain any coded one-qubit gate, and a specific two-qubit gate, the controlled NOT (CNOT).

We now show how one-qubit gates are performed on a single three-spin block. We write the Heisenberg hamiltonian as  $H_{ij} = J \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j$  between spins  $i$  and  $j$ . Hamiltonian  $H_{12}$  generates a rotation  $U_{12} = \exp(i\hbar^{-1} \int \mathbf{J} \mathbf{S}_1 \cdot \mathbf{S}_2 dt)$  which is just a  $z$ -axis rotation (in Bloch-sphere notation) on the coded qubit, while  $H_{23}$  produces a rotation about an axis in the  $x$ - $z$  plane, at an angle of  $120^\circ$  from the  $z$ -axis. As simultaneous application of  $H_{12}$  and  $H_{23}$  can generate a rotation around the  $x$ -axis, three steps of one-dimensional (1D) parallel operation (defined in Fig. 1a) permit any one-qubit rotation, using the classic Euler-angle construction. In serial operation, we find numerically that four steps are always adequate when only nearest-neighbour interactions are possible (for example, the sequence  $H_{23} - H_{12} - H_{23} - H_{12}$  shown in Fig. 2a, with suitable interaction strengths), while three steps suffice if interactions can be turned on between any pair of spins (for example,  $H_{23} - H_{12} - H_{13}$ ; Fig. 2b).

We have performed numerical searches for the implementation of two-qubit gates using a simple minimization algorithm. Much of the difficulty of these searches arises from the fact that while the four basis states  $|0_L, 1_L\rangle|0_L, 1_L\rangle$  have total spin quantum numbers  $S = 1$ ,  $S_z = +1$ , the complete space with these quantum numbers for six spins has nine states, and exchanges involving these spins perform rotations in this full nine-dimensional space. So, for a given sequence, such as that depicted in Fig. 2c, we consider the resulting unitary evolution in this nine-dimensional Hilbert space as a function of the interaction times  $t_1, t_2, \dots, t_N$ . This unitary evolution can be expressed as a product  $U(t_1, \dots, t_N) = U_N(t_N) \dots U_2(t_2) U_1(t_1)$ , where  $U_n(t_n) = \exp(it_n H_{i(n),j(n)}/\hbar)$ . The objective of the algorithm is to find a set of interaction times such that the total time evolution describes a CNOT gate in the four-dimensional logic subspace  $U(t_1, \dots, t_N) = U_{\text{CNOT}} \oplus A_5$ . The matrix  $A_5$  can be any unitary  $5 \times 5$  matrix (consistent with  $U$  having a block diagonal form). The efficiency of our search is considerably improved by the use of two invariant functions



**Figure 1** Possible layouts of spin-1/2 devices. **a**, One-dimensional layout. We consider two different assumptions about how the exchange interactions can be turned on and off in this layout: (1) at any given time each spin can be exchange-coupled to at most one other spin (we refer to this as “serial operation” in the text); (2) all exchange interactions can be turned on simultaneously between any neighbouring pair of spins in the line shown (“1D parallel operation”). **b**, Possible two-dimensional layout with interactions in a rectangular array. We imagine that any exchange interaction can be turned on between neighbouring spins in this array (“2D parallel operation”). Other arrangements are possible, but those shown here should be representative of the constraints that will be faced in actual device layouts.

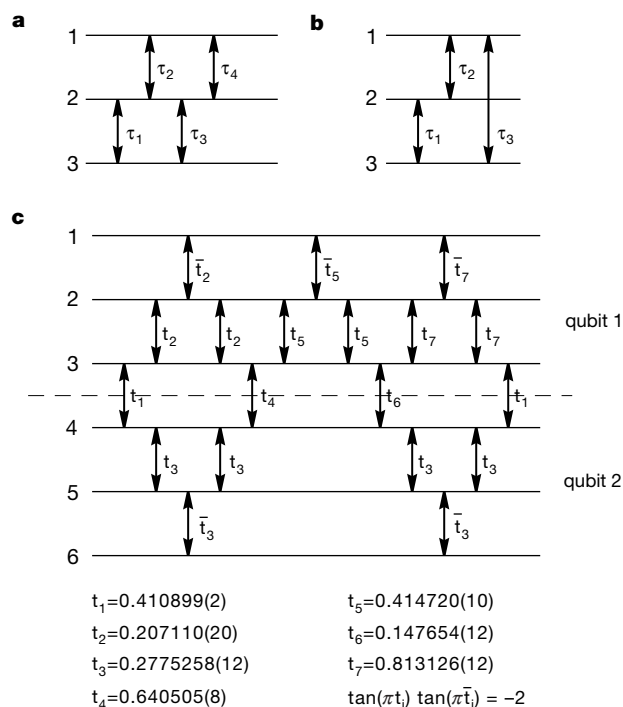
$m_{1,2}(U)$  identified by Makhlin<sup>19</sup>, which are the same for any pair of two-qubit gates that are identical up to one-qubit rotations. It is then adequate to use an algorithm that searches for local minima of the function  $f(t_1, \dots, t_N) = \sum_i (m_i(U_{\text{CNOT}}) - m_i(U(t_1, \dots, t_N)))^2$  with respect to  $t_1, \dots, t_N$  ( $m_i$  is understood only to act on the  $4 \times 4$  logic subspace of  $U$ ). Finding a minimum for which  $f = 0$  identifies an implementation of CNOT (up to additional one-qubit gates, which are easy to identify<sup>19</sup>) with the given sequence  $(i(n), j(n))_n$ ,  $i(n) \neq j(n)$  of exchange gates. If no minimum with  $f = 0$  is found after many tries with different starting values (ideally mapping out all local minima), we have strong evidence (although not a mathematical proof) that the given sequence of exchange gates cannot generate CNOT.

The optimal serial-operation solution is shown in Fig. 2c. We note that this solution happens to involve only nearest neighbours in the 1D arrangement of Fig. 1a. The circuit layout shown has a high degree of symmetry; however, it does not appear possible to give the obtained solution in a closed form. (Any gate sequence involving non-nearest neighbours can be converted to a local gate sequence by swapping the involved qubits, using the SWAP gate, until they are close; but here the minimal solution found does not require such manipulations.) We have also found (apparently) optimal numerical solutions for parallel operation mode. For the 1D layout of Fig. 1a, the simplest solution found involves 8 clock cycles with just  $8 \times 4$  different interaction-time parameters ( $H_{12}$  can always be zero in this implementation). For the 2D parallel mode of Fig. 1b, a solution was found using just 7 clock cycles ( $7 \times 7$

interaction times). The interaction parameters for these solutions are available from the authors on request.

It is worthwhile to give a complete overview of how quantum computation would proceed in the present scheme. It should begin by setting all the computational qubits to the  $|0_L\rangle$  state. This state may be obtained using the exchange interaction: if a strong  $H_{12}$  is turned on in each coded block and the temperature made lower than the strength  $J$  of the interaction, these two spins will equilibrate to their ground state, which is the singlet state. The third spin in the block should be in the  $|1\rangle$  state, which can be achieved by also placing the entire system in a moderately strong magnetic field  $B$ , such that  $k_B T \ll g\mu_B B < J$ . Then, computation can begin, with the one- and two-qubit gates implemented according to the schemes mentioned above. For the final qubit measurement, we note that determining whether the spins 1 and 2 of the block are in a singlet or a triplet suffices to perfectly distinguish  $|0_L\rangle$  from  $|1_L\rangle$  (again, the state of the third spin does not enter). Thus, for example, the a.c. capacitance scheme for spin measurement proposed by Kane<sup>3</sup> is directly applicable to the coded-qubit measurement.

There are several issues raised by this work that deserve further exploration. The  $S = 1/2$ ,  $S_z = +1/2$  three-spin states that we use are a subspace of a decoherence-free subsystem that has been suggested for use in quantum computing by exchange interactions<sup>10,16</sup>. Use of this full subsystem, in which the coded qubit can be in any mixture of the  $S_z = +1/2$  and the corresponding  $S_z = -1/2$  states, would offer immunity from certain kinds of interactions with the environment, and would not require any magnetic field to be present, even for initialization of the qubits.



**Figure 2** Circuits for implementing single-qubit and two-qubit rotations using serial operations. **a**, Single-qubit rotations by nearest-neighbour interactions. Four exchanges (double-headed arrows) with variable time parameters  $\tau_i$  are always enough to perform any such rotation; one of the two possible layouts is shown. **b**, Non-nearest neighbour interactions. Only three interactions are needed; one of the possible layouts is shown. **c**, Circuit of 19 interactions that produce a CNOT between two coded qubits (up to one-qubit gates before and after). The durations of each interaction are given in units such that for  $t = 1/2$  the rotation  $U_{ij} = \exp(i/t \mathbf{S}_i \cdot \mathbf{S}_j / \hbar)$  is a SWAP, interchanging the quantum states of the two spins  $i, j$ . The  $\bar{t}_i$  parameters are not independent, but are related to the  $t$  values as indicated. The uncertainty of the final digits of these times is indicated in parentheses. With these uncertainties, the absolute inaccuracy of the matrix elements of the two-qubit gate rotations achieved is no greater than  $6 \times 10^{-5}$ . Further fine-tuning of

these time parameters would give the CNOT to any desired accuracy. In a practical implementation, the exchange couplings  $J(t)$  would be turned on and off smoothly; then the time values given here provide a specification for the integrated value  $\int J(t) dt$ . The functional form of  $J(t)$  is irrelevant, but its integral must be controlled to the precision indicated. The numerical evidence is very strong that the solution shown here is essentially unique, so that no other choices of these times are possible, up to simple permutations and replacements  $t \rightarrow 1 - t$  (we note that for the Heisenberg interaction adding any integer to  $t$  results in the same rotation). The results also strongly suggest that this solution is optimal: no one of these 19 interactions can be removed, and no other circuit layout with fewer than 19 has been found to give a solution. We have also sought, but not found, shorter implementation of other two-qubit gates like  $\sqrt{\text{SWAP}}$  (refs 2, 5).



In this modified approach, the implementation of one-qubit gates is unchanged, but the CNOT implementation must satisfy additional constraints—the action of the exchanges on both the  $S = 1$  and the  $S = 0$  six-spin subspaces must be considered. As a consequence, implementation of CNOT in serial operation is considerably more complex; our numerical studies have failed to identify an implementation (even a good approximate one) for sequences of up to 36 exchanges (compare with 19 in Fig. 2c). On the other hand, we have found implementations using 8 clock cycles for 1D and 2D parallel operation (again for the 1D case  $H_{12}$  can be zero), so use of this larger Hilbert space may well be advantageous in some circumstances.

Finally, we note that further work is needed on the performance of quantum error correction within this scheme. Our logical qubits could be used directly within the error correction codes that have been shown to produce fault-tolerant quantum computation<sup>20</sup>. Spin decoherence would primarily result in ‘leakage’ errors, which would take our logical qubits into states of different angular momentum (for example,  $S = 3/2$ ). Our preliminary work indicates that, with small modifications, the conventional error correction circuits will not cause uncontrolled propagation of leakage error. In addition, the general theory<sup>8,10,20,21</sup> shows that there exist sequences of exchange interactions that directly correct for leakage by swapping a fresh  $|0_L\rangle$  into the coded qubit if leakage has occurred, and doing nothing otherwise; we have not yet identified numerically such a sequence. If fast measurements are possible, teleportation schemes could also be used in leakage correction.

The present results offer an alternative route to the implementation of quantum computation. The trade-offs are clear: for the price of a factor of three more devices, and about a factor of ten more clock cycles, the need for stringent control of magnetic fields applied to individual spins is dispensed with. We hope that the flexibility offered by these results will make easier the hard path to the implementation of quantum computation in the laboratory. □

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## Kondo physics in carbon nanotubes

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The connection of electrical leads to wire-like molecules is a logical step in the development of molecular electronics, but also allows studies of fundamental physics. For example, metallic carbon nanotubes<sup>1</sup> are quantum wires that have been found to act as one-dimensional quantum dots<sup>2,3</sup>, Luttinger liquids<sup>4,5</sup>, proximity-induced superconductors<sup>6,7</sup> and ballistic<sup>8</sup> and diffusive<sup>9</sup> one-dimensional metals. Here we report that electrically contacted single-walled carbon nanotubes can serve as powerful probes of Kondo physics, demonstrating the universality of the Kondo effect. Arising in the prototypical case from the interaction between a localized impurity magnetic moment and delocalized electrons in a metallic host, the Kondo effect has been used to explain<sup>10</sup> enhanced low-temperature scattering from magnetic impurities in metals, and also occurs in transport through semiconductor quantum dots<sup>11–18</sup>. The far greater tunability of dots (in our case, nanotubes) compared with atomic impurities renders new classes of Kondo-like effects<sup>19,20</sup> accessible. Our nanotube devices differ from previous systems in which Kondo effects have been observed, in that they are one-dimensional quantum dots with three-dimensional metal (gold) reservoirs. This allows us to observe Kondo resonances for very large electron numbers ( $N$ ) in the dot, and approaching the unitary limit (where the transmission reaches its maximum possible value). Moreover, we detect a previously unobserved Kondo effect, occurring for even values of  $N$  in a magnetic field.

Each of our devices<sup>21</sup> contains a metallic single-walled nanotube with gold source and drain contacts and a substrate gate contact, as shown in Fig. 1a. Several measures are taken to optimize electrical contact between the gold and the nanotubes (see Methods). The resulting two-terminal linear-response conductance  $G$  ranges up to more than  $3e^2/h$  (resistance  $8\text{ k}\Omega$ ) at room temperature, where  $h$  is Planck’s constant and  $e$  is the electronic charge. We believe this is the highest reported yet for a single-walled tube<sup>22</sup>. This implies that the transmission probability  $P$  of each contact can reach about 0.9 (the maximum theoretical conductance of a metallic tube is  $4e^2/h$ ), demonstrating that nearly ideal contacts are achievable between a normal metal and a molecule. Meanwhile, the variation of  $P$  from device to device allows the investigation of different transport regimes.

For  $P \ll 1$ , as is well established<sup>2,3</sup>, Coulomb blockade occurs at low temperature  $T$  and the device behaves as a one-dimensional (1D) quantum dot<sup>23</sup>. The values of the charging energy,  $U$  (the average interaction between two electrons), and the average level spacing,  $\Delta E$ , found for this type of device are as expected for a tube