Efficient quantum computing on low temperature spin ensembles.

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A new scheme is proposed which will permit electron spin resonance pulse techniques to be used to realize a quantum computer with a 100 qbits, or more. The computation is performed on effective pure states which correspond to off-diagonal blocks of the density matrix. Described is a scheme which very efficiently performs the preparation stage and which permits “pseudo-projective measurement” to be made on the output. With such measurements all members of the ensemble remain coherent.

The demonstration by Shor (1) that a quantum computer can factor large numbers in a time which goes as a polynomial rather than the exponential of the number of digits has electrified interest in quantum computers.

In a quantum calculation an initial state $|\vec{0}\rangle = |0\ldots0\rangle$ is acted upon by a unitary transformation $U$ which reflects the calculation and which gives a result $|R\rangle = U|\vec{0}\rangle$. In order that the calculation make sense $|\vec{0}\rangle$ must be a perfectly specified state and it must be possible to determine $|R\rangle$. The designer of a quantum computer is immediately faced by three non-trivial problems. First, since $|\vec{0}\rangle$ is some particular state, the entropy is zero and the system must be cooled to absolute zero. Second, the fundamental tenants of quantum mechanics insist a given quantum state $|R\rangle$ cannot be determined accurately by a single measurement. Repeated calculation cycles are necessary even before concerns of the signal to noise are addressed. Third, it is necessary to read the individual qbits.

Cirac and Zoller (2) have suggested a physical realization of a quantum computer based on linear ion traps, however even supporters (3) of such devices as a proof-of-principle for quantum computing concede that these will never make a useful computer. Ingenious schemes for performing computations using standard liquid room temperature nuclear magnetic resonance (NMR) techniques has been introduced by Gershenfeld and Chuang (4) and Cory et al (5). Again there is a serious scaling problem and it is difficult to imagine
a computer with much more than 10 qbits. As pointed out by Warren (6) in a ensemble of $10^{23}$ computing molecules at room temperature there is essentially zero probability of finding a single example of any state designated to be $|\overline{0}\rangle$ for a 100 qbit register.

An efficient $\sim 100$ qbit ensemble spin computer requires an almost complete polarization of the spins and almost inevitably low temperatures, electronic spins, and the solid state. Commercial pulse electron spin resonance (ESR) spectrometers operating at $\sim 100$ GHz are available. For spins with $g = 2$ this corresponds to a field of 3T. At a temperature of 1K, $e^{\hbar\omega_0/k_B T} \sim 10^{-4}$ so for a 100 qbit computer a large fraction of the computing molecules will in the ground state designated to be $|\overline{0}\rangle$. A basic figure of merit $Q = \omega_0 \tau$ is the ratio of $\omega_0$ the transition frequency ($1/\omega_0$ is the shortest switching time) and $\tau$ the coherence time. As a general rule, light elements have the longest $\tau$, e.g., Li (7) or organic free (8) radicals have $g$-factors very close to 2.00 and very long $T_1 \sim 10^{-3}$s times, implying $Q$ values as large as $10^9$ in the solid state. For comparison DiVincenzo (9) gives $Q = 10^7$ for NMR and $Q = 10^{13}$ for ion traps. However these latter figures are misleading since the implementation of quantum gates requires individual qbits to be addressed. Both linear traps and liquid NMR are very slow because $\Delta\omega_0$ the differences in frequencies are very small. In order to avoid this difficulty, it is envisaged here that each qbit comprises two spins. Rare earth, i.e, 4f ions such as Dy and Er often have $g \sim 6 - 7$ but somewhat shorter $T_1$ times. It is imagined that, e.g., a free radical plus 4f ion bi-spin unit would be a basic building block with the 4f ion always in its very long lived ground state. The intra-unit exchange interaction between this 4f moment and the computing spin via differing bridge molecules would provide large shifts from $g = 2$. The inter-unit exchange provides the necessary interactions for qbit manipulation. The technology envisaged is that of molecular magnetism which has been much developed (10) over the past decade or so.

The detailed realization of such a machine is not the principal concern here. The questions asked assume that such physical systems can be engineered. Consider the following structure for a register of a quantum computer: There is a alternating chain of $N$ computing spins which have distinct resonance frequencies $\omega_n$ even in the absence of interactions. To simplify the translation to the computational problem, for each computing spin the notation $|\uparrow\rangle = |1\rangle$ and $|\downarrow\rangle = |0\rangle$ will be adopted and it will be assumed that $|0\rangle$
corresponds to the ground state. Due to interactions, each spin sees its two neighbours, so that each resonance at $\omega_n$ is split into four addressable (well separated) lines with frequencies $\omega_n^{00}$, $\omega_n^{01}$, $\omega_n^{10}$ and $\omega_n^{11}$ as the neighbours are 00, 01, 10 and 11 respectively. Here 10 means that the spin $n-1$ is 1 while the $n+1$ spin is 0. Because of the alternation of the interaction $\omega_n^{01} \neq \omega_n^{10}$. A $\pi$ pulse at any one of these frequencies amounts to the execution of a generalized Toffoli gate, i.e., such a pulse, e.g., at the frequency $\omega_n^{10}$ will flip the $n$th spin if, and only if, its neighbors are “1” and “0”. This gate along with arbitrary spin rotations can be effected by standard pulse resonance technique and permit all quantum gates to be constructed. This has been adequately dealt with by others (4,5,11,12) and will not be described here.

Even with the considerable advantage of low temperature electronic spin systems there remain two very important problems in using finite temperature ensembles for quantum computing. First, even if they are relatively few, it is still necessary to eliminate the signals from molecules not in the ground state $|0\rangle$. The result $|R\rangle = U|0\rangle$ will be a linear superposition of many states and the weight of a given state in $|R\rangle$ will very often be much smaller than the weight of unwanted signals from other members of the thermodynamic ensemble. Necessary is a preparation (or final averaging) procedure in order that the observable signal originates only from the computation pure state $|0\rangle$. Most of the procedures suggested to date (4,5,12,13) are for small high temperature ensembles. They typically become exponentially long for large registers negating the advantage of performing a quantum computation. Potentially non-exponential schemes do exist (13) although no specific implementation has been suggested for a low temperature ensemble. The principle of these earlier techniques is quite different from that proposed here. The usual idea is to reshape the entire density matrix to be an effective pure state of the form $(1/N) + p|\bar{0}\rangle < \bar{0}|$, while here the aim is to create off-diagonal blocks $|\sigma\rangle |\bar{0}\rangle < < \bar{0}| - \sigma|$, $\sigma = 0, 1$ which correspond to an ESR signature from a “signal” spin reflected by $\sigma$. The second problem has to do with projective measurements. Quantum computations typically involve measuring the state of $|R\rangle$ and, after the principles of quantum mechanics, this involves a random projection. If such measurements were possible using NMR, or ESR, pulse techniques this would leave different members of the ensemble in randomly different
states. A remedy for this difficulty has been suggested (4), however it remains the case that such perfectly projective measurements do not exist in the repertoire of NMR, or ESR, pulse techniques, although a clever such scheme does exist for ion traps (14). It is always possible to imagine deterministic equivalent of a given quantum algorithm although no explicit such scheme has yet to be presented of the factoring problem. Here is proposed a general method by which to perform “pseudo-projective” measurements and which permit the existing algorithms to used directly.

The basic idea of the new preparation scheme is to perform a simple quantum computation on all the states of the ensemble which permits the free induction ESR resonance signal to be reduced to that coming from the state $|0\rangle$. A signal spin, the ESR of which is monitored, is added as the first spin of a $N+1$ spin register, thus if $|A\rangle$ is an arbitrary ($N$ spin) state, the state of the whole register is $|\sigma\rangle|A\rangle$, $\sigma = 0, 1$. The equilibrium ensemble gives no (free induction decay) ESR signal. A signal corresponding the transitions $|\sigma\rangle \leftrightarrow |-\sigma\rangle$, for states of the form $|\sigma\rangle|0A\rangle$, is turned “on” by $\pi/2$ pulse at $\omega^0_1$. The standard “spin echo” technique comprises a sequence of gradient-$\pi$-gradient pulses (15). A field gradient applied to the sample causes the spins on different molecules to precess at different rates and this destroys the ESR signal, however the signal is recovered following a time conjugating $\pi$ pulse if an identical gradient pulse is applied to re-focus the spins. The principle of the preparation stage is, between gradient pulses, to perform a computation on all the states of the ensemble which time conjugates the signal spin if, and only if, the state $|A\rangle = |0\rangle$. Conceptually the simplest scheme would be to consider the signal spin as a separate single bit register $s_1$ and perform a quantum calculation equivalent to the pseudo code

$$\text{If } A = 0, s_1 = s_1 + 1 \pmod{2}$$

The result of a computation is $|\sigma\rangle|R\rangle = |\sigma\rangle U|0\rangle$ where $U$ is the relevant unitary transformation which has no effect on the signal spin. Projective measurements are replaced by what might be called “pseudo-projective” reversible equivalents. The principle (again the practice is somewhat different) is similar to the preparation stage. The presence of a state $|\sigma\rangle|S_0\rangle \equiv |\sigma\rangle|\sigma_2, \sigma_3 \ldots \sigma_N\rangle$ is detected by a signal spin ESR by performing the equivalent of a calculation on all states $|\sigma\rangle|A\rangle$, between two gradient
pulses, which reverses the signal spin if, and only if, \( A = S_0 \), i.e., the pseudo code is:

\[
\text{If } A = S_0, \ s_1 = s_1 + 1 \pmod{2}
\]

The same pulse sequences in the opposite order return the system to the state before the interrogation process and hence this reading process is reversible. Usually the result cannot be determined by a single such measurement and measuring every amplitude takes an exponentially long time. Described here are routines which take, at the most, \( \sim N^2 \) pulses to read the result for either the Shor or Grover (16) algorithms.

While the advantage of a reversible “read” process are considerable, there is also a major potential disadvantage over projective measurements. After a perfect projective measurement (if such a thing really exists), of, e.g., spin orientations corresponding to the set of operators \( \hat{\sigma}_n^z \); \( n = 1, N + 1 \), the system finds itself in an eigenstate \( \prod_{n=1}^{N+1} |\sigma_n^z > \) of these operators. The weight of this state in original wavefunction is reflected not in the strength of the associated “signal” but rather by the probability of finding the particular signature corresponding to the set \( \{\sigma_n^z\} \). In a straightforward reversible measurement of a quantum state the strength of the signal must be proportional to the weight squared of the particular state. In a \( N + 1 \) qbit register, and the worst case when all states have roughly the same weight, the strength of a reversible measurement is \( \sim 2^{-N} \) as is the probability of a particular result of a projective measurement. An efficient quantum algorithm is explicitly constructed to have a \( |R > \) with much less quantum entanglement, however this can remain a difficulty for reversible reads. In this regard Grover’s algorithm presents no problem since the answer dominates the register to be measured. The Shor scheme requires more discussion which will be taken up again below.

The first step is to describe an efficient scheme which reduces the ESR response of the signal spin to that coming from the two computational \( |\sigma > |\bar{0} > \) states. This uses no ancilla spins other than the signal spin. As described above, the signal comes only from states of the form \( |\sigma > |0A > \). After a first gradient pulse, are \( \pi \) pulses at the frequencies \( \omega_{n}^{10} ; n = 2, N \). A second less selective \( \pi \) pulses sequence, which only “looks left”, has frequencies \( \omega_{n}^{0X} ; n = N, 2 \) where \( X = 0 \) or \( 1 \). (It is implied that pulses are applied at, or cover, both \( \omega_{n}^{00} \) and \( \omega_{n}^{10} \).) Last is a \( \pi \) pulse at \( \omega_{1}^{1} \). For the computational states the net effect is \( |1 > |0 \ldots 00 > \rightarrow |0 > |1 \ldots 10 > \) and \( |0 > |0 \ldots 00 > \rightarrow |1 > |1 \ldots 10 > \). If the last
spin was reversed this would be a complete time conjugation, however this is not necessary since both states de-phase in the same manner for a given molecule and so a second identical gradient pulse does re-focus the spins and the ESR of the signal spin is recovered.

For the non-computational states there are two cases. If the first “1” occurs on the third spin, the net effect is $|1 > |01A >\rightarrow |1 > |00A'' >$ and $|0 > |01A >\rightarrow |1 > |10A'' >$ where $A''$ is some unique mapping of $A$. Generally the first “1” occurs after $M$ zeros $|1 > |0\ldots001A >\rightarrow |0 > |1\ldots100A'' >$ while $|0 > |0\ldots001A >\rightarrow |1 > |1\ldots110A'' >$ where ... replaces a series of digits “1”. In all cases the resulting states differ at the qbit positioned two places before the “A” and as a result the final states dephase differently during the second gradient pulse and the ESR signal is not recovered. In addition, for the first case, the signal spin is not properly conjugated.

This preparation stage permits the extraction of the result of the quantum computation on the pure states $|\sigma > U |\overline{0} >$. The transformation $U$ does not change the signal spin ESR. The weight of any given state $|\sigma_2\ldots\sigma_N >$ in $U |\overline{0} >$ might be determined by another gradient-$\pi$-gradient sequence, however it would take an exponentially large time $\sim 2^N$ to measure the amplitude of every state. There are probably as many strategies for rendering practical such a scheme as there are quantum algorithms (so finding them is not a big task as of writing).

The Shor routine searches for the period $r$ of $f(a) = x^a \text{ Mod} N$ where $N$ is a number to be factored and $x < N$ is randomly chosen. The result is $|R >=(1/\sqrt{r}) \sum_{p=0}^{r-1} |pT > |F_p >$, and the sought for $r = w/T$ where $w$ corresponds to the size of the registers and where $T$ is the period, i.e., the smallest non-trivial number in the first register. The (normalized) $|F_p >$ need not be read. This smallest number has the largest quantity, $N$, of leading zeros, i.e., it is of the form $|\sigma > |0\ldots0A >\equiv |s_1 = \sigma > |S_\beta = 0\ldots0 >, |A >, >, >, \beta$. The pseudo-code which conjugates the signal spin for such a state is

If $S_\beta = 0, s_1 = s_1 + 1 \text{ (mod 2)}$

This program line is run with $N = 1, 2\ldots$ until it fails, with $N = N_0$, to conjugate the signal spin. The last such pulse sequence is then run backwards to recover the result before the failure. The smallest number then begins $|\sigma > |0\ldots01A' >$ with $N_0 - 1$ zeros and the search is continued for the smallest $A'$, etc.
Again a more efficient equivalent process is possible. The $|\sigma > |0A>$ will exist if a significant signal at $\omega^0_1$ is observed. Assuming this is the case, a finite amplitude for the states $|\sigma > |00A>$ is sought. First the signal from the $|1A>$ states is “killed” by a $\pi/2$ pulse at $\omega^1_1$. The remaining task is relatively simple since it is known where the first “error” will occur. As above the process starts with a gradient pulse, followed by $\pi$ pulses at $\omega^1_1$ and $\omega^0_1$. The net changes are $|1 > |01A >\rightarrow |1 > |11A >$, $|1 > |00A >\rightarrow |0 > |00A >$, $|0 > |00A >\rightarrow |1 > |00A >$ and $|0 > |01A >\rightarrow |1 > |01A >$, i.e., this performs the necessary conjugation on the signal spin. The final gradient pulse will restore the ESR signal from the states $|\sigma > |00A>$ while leaving the states originating from $|\sigma > |01A>$ with the signal turned off.

The generalization to $|\sigma > |0 . . . 0A>$, where there are $N$ zeros, contains the $\pi$ sequences $\omega_{n}^{10}$, $n = 2, N - 1$; $\omega_{N}^{11}$ followed by $\omega_{n}^{10}$, $n = N - 1, 2$ which resets the state $|1 > |0 . . . 00A>$ while having the effect $|1 > |0 . . . 01A >\rightarrow |1 > |11 . . . 11A>$ but leaving the states $|0 > |0 . . . 00A>$ and $|0 > |0 . . . 01A >$ unchanged. Clearly the sandwiching gradient pulses destroy all but the signal from $|\sigma > |0 . . . 0A>$. The generalization to the search for other specific states is evident.

The signal corresponding to $(1/\sqrt{r})|T>$ is reduced by a factor $\sim 1/r$ where $r < N$, $N$ being the number to be factorised. It is possible to detect $\sim 10^5$ equivalent spins, so if $r > 10^{18}$ even for a $10^{23}$ element ensemble the signal for a $\sim 100$ qbit register will become unreadable. However a pseudo-collapse of the wavefunction can be accomplished by changing, a little, Shor’s algorithm. The usual calculation, $U$, causes $|a > |b >\rightarrow |a > |x^a b>$, where again $x$ is a randomly chosen $c$-number less than $N$. The pseudo-collapse is accomplished by modifying the second register so that the effect of $U$ is $|a > |1 > +|x > +|x^2 > + . . . +|x^n > + . . . +|x^s >\rightarrow |a > |x^a > +|x^{a+1} > +|x^{a+2} > + . . . +|x^{a+n} > + . . . +|x^{a+s} >$ where $s < r$. This modification can be performed efficiently but requires an extra register. Following an initial failure to read $|T>$ with $s = 0$, a second calculation with $s \sim 10^{18}$ must succeed.

For the Grover algorithm the answer dominates $U|\overline{0}>$. First the signal spin is observed. There will be a single strong line which determines the leading digit in $U|\overline{0}>$. Imagine, e.g., that this implies $U|\overline{0} >\sim |1A>$, using the method of the previous few para-
graphs the amplitude of $|11A >$ is determined. If, e.g., this is small then the state begins $|10A >$ and the last interrogation sequence is run backwards to recover the amplitude for this state and then $|101A >$ sought for, etc. (An alternative but somewhat more “rough and ready” method is to apply a small angle, say $\pi/20$, pulse to all spins. This will turn “on” an ESR signal which corresponds to the desired state, e.g., if $U|0 > \sim |10100010 \ldots >$ then the $n = 2$ spin will give a free induction signal at the two positions $\omega_2^{X1}$ and then at $\omega_3^{11}, \omega_4^{00}, \omega_5^{10}, \ldots$)

Error correction represents an interesting challenge. In order to remove the entropy generated by errors, such schemes (17) use projective measurements in an essential fashion. Replacing these by the pseudo-projective equivalents would cause the signal strength to fall off exponentially. However it is not hard to find alternative means by which to perform the correction using conditional unitary transformations. The entropy must be carried away by ancillary qbits and then removed by cooling. A full discussion of these possibilities is too lengthy to be reproduced here.

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REFERENCES AND NOTES

1. P. Shor, In *Proceedings of the 35th Annual Symposium on Foundations of Computer Science*, (IEEE Computer Society, Los Alamitos, CA, 1994).
2. J. I. Cirac and P. Zoller, *Phys. Rev. Lett.* **74** 4091-4094 (1995).
3. D. Beckman, A. Chari, S. Devabhaktuni and J. Preskill, *Phys. Rev. A* **54**, 1034-1063 (1996).
4. N. A. Gershenfeld and I. L. Chuang, *Science* **275** 350-356 (1997).
5. D. G. Cory, A. F. Fahmy and T. F. Havel *Proc. of the 4th Workshop on Physics and Computation*, (New England Complex Systems Institute, Boston, MA 1996).
6. W. S. Warren, *Science*, **277** 1688 (1997).
7. R. Jones, J. A. Howard, H. A. Joly, P. P. Edwards and R. J. Singer, *Mag. Resonance in Chem.*, **33** S98 (1995).
8. See e.g., J. Veciana, J. Cirujeda, C. Rovira and J. Vidal-Gancedo, *Adv. Mat.* **7**, 221 (1995).
9. D. P. DiVincenzo, *Phys. Rev. A* **51** 1015-1022 (1995).
10. see, e.g., O. Kahn, *Molecular Magnetism*, (VCH Publishers, Inc., 220 East 23rd Street, New York, NY 10010-4606, 1993), D. Gatteschi, *Current Opinion in Solid State and Mat. Science*, **1**, 192 (1996).
11. S. Lloyd, *Science*, **261** 1569 (1993); see also *Science*, **263** 695 (1994).
12. D. G. Cory, M. D. Price and T. F. Havel, quant-phys/970900, (1997).
13. E. Knill, I Chuang and R. Laflamme, quant-phys/19706053, (1997).
14. W. Nagourney et al., *Phys. Rev. Lett.* **56**, 2797 (1986); J. C. Bergquist et al. *Phys. Rev. Lett.* **56**, 1699 (1986); T. Sauter et al., *Phys. Rev. Lett.* **56**, 1696 (1986).
15. See e.g., C. P. Slichter, *Principles of Magnetic Resonance*, (3rd. Ed., Springer-Verlag, Heidelberg Germany, 1989).
16. L. K. Grover, *Phys. Rev. Lett.*, **79**, 325 (1997).
17. See, e.g., D. P. DiVincenzo and P. W. Shor, *Phys. Rev. Lett.* **77**, 3260 (1996) and references therein.