I. Introduction

Quantum computing–the manipulation of a quantum mechanical system to do information processing– has attracted considerable recent attention, largely triggered by Shor's recently proposed algorithm for finding prime factors in polynomial instead of exponential time. The importance of this problem in communications and cryptography has also led to numerous attempts to realize quantum computers, mostly in exotic systems such as trapped ions. Very recently, several research groups have proposed the use of bulk samples at room temperature to do quantum computing–specifically, nuclear magnetic resonance (NMR) of molecules in a room temperature solution.

The research in this proposal addressed a variety of *fundamental* experimental and theoretical issues associated with bulk quantum computing. The goal is to determine if, in fact, there exist any bulk systems which will ultimately be reasonable candidates for quantum computing. For example, we have recently shown that solution NMR quantum computers, while an elegant concept, suffer from several limitations which are independent of any assumed computing algorithm and are not present in other potential implementations. In fact, if today's understanding of quantum mechanics and statistical mechanics as applied to magnetic resonance is correct (and there is no experimental evidence to the contrary), solution NMR quantum computing will *never* be scalable to a useful number of bits. However, these limitations do not apply to many other potential realizations of quantum computers, which thus seem more credible. Our experimental work will concentrate on what we believe to be the most promising bulk systems–electron spin resonance (ESR) of systems with multiple radicals, and shaped-pulse laser spectroscopy (using high resolution shaping techniques developed in this laboratory) of mixed molecular crystals. Our goal was to determine what molecules would be optimal sources of qubits, and to estimate the maximum feasible complexity and computational speed.

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14. ABSTRACT

Solution NMR quantum computers are fundamentally limited to small numbers of qubits because energy differences are small compared to kT, and the logic gates that can be created are extremely slow (ca. 100 ms) (see W. S. Warren, Science 277, 1688-1689 (1997)). We proposed to explore the technological and architecture issues associated with two other classes of bulk quantum computing systems: ESR and optical impurity molecules in a dilute lattice. In the ESR case, we synthesized specific molecules that were capable in principle of producing single logic gates. We also dramatically advanced the technology for manufacturing phase coherent femtosecond laser pulse sequences, a critical component of any architecture which could achieve meaningful speedup. Finally, we explored the theoretical issues associated with specific classes of decoherence.

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II. Results

A. ESR Quantum Computing

Electron spin resonance (ESR) shares many important characteristics with NMR. The Hamiltonian is quite simple (consisting essentially of variations in the g factor and hyperfine plus dipolar couplings). Variations in the g factor have the same mathematical form as chemical shifts; hyperfine and electron-electron dipolar couplings are bilinear, and in the limit that they are smaller than the g factor variations, the relevant portion is identical to the first-order J coupling in solution NMR. Relaxation times can be quite long at low temperatures (milliseconds); 90^o and 180^o pulses can be have durations of a few tens of ns. Pulsed methods, including two-dimensional experiments, are possible with commercially available spectrometers, including the 10 GHz Bruker spectrometer installed at Princeton.

From a quantum computing perspective, ESR has several critical advantages. The most important advantage is that the gyromagnetic ratio of the electron is 650 times greater than that of the proton, hence the transition frequencies are that much higher. This means, for example, that ESR spectroscopy in modern superconducting magnets (resonance frequencies around 300GHz) can achieve $h\nu >> kT$ at liquid helium temperatures. For transitions with $h\nu >> kT$ the initial state can be prepared essentially without loss, no matter how many systems are coupled. In addition, the couplings (hence the potential speed) are vastly larger than J couplings in solution NMR–in the experiments we will describe below, couplings of 10^7-10^8 Hertz will be common. Finally, only unpaired spins give ESR spectra, so solid state experiments are no longer so difficult–to a first approximation, a mixed crystal with dilute radicals in a radical-free host can be treated as if the host crystal did not exist.

The most common form of ESR spectroscopy uses the hyperfine interaction to produce sharp multiplets for unpaired spins which are delocalized over several atoms (for example, aromatic radicals). In this case N nuclear spins and one electron spin produce 2^{N+1} energy levels, and highly structured ESR spectra are readily produced. However, in this case all of the 2^N different nuclear spin states with the electron spin up lie within a small energy band, and the scaling problem is identical to NMR–it is essentially impossible to cool the system enough to put everything in the ground state. Instead, we propose to study systems with multiple radicals in the same molecule. The radicals are coupled by the dipole-dipole interaction (if they are highly separated) or by an exchange interaction if they are nearer. Now all of the 2^N energy levels for N coupled radicals are separated by electron spin flips, and cooling to the ground state is feasible. Thus ESR provides a system which *is* in principle scalable to useful numbers of bits, and which also can execute logic gates at a reasonable speed.

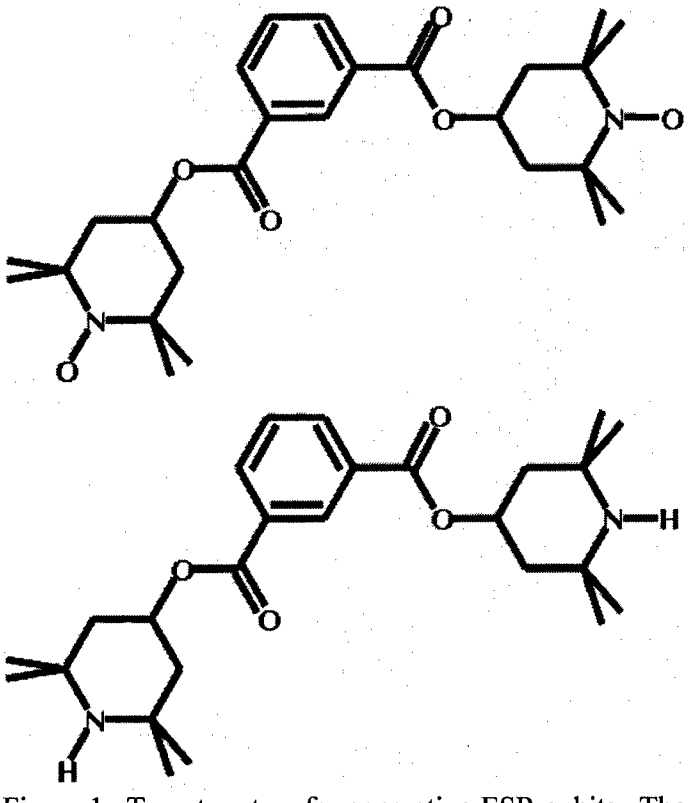

Figure 1. Target system for generating ESR qubits. The molecule at the top has nitroxide radicals coupled by dipole-dipole interactions. It was embedded in the diamagnetic host matrix corresponding to the molecule at the bottom.

The top of Figure 1 shows a linked pair of spin-

1/2 nitroxides which we synthesized, then embedded in a diamagnetic host (the molecule at the bottom). This is an excellent example of an effective two-qubit ESR system, with a very large coupling matrix element between the two qubits. The easiest way to verify this is to do ESR spectroscopy (Figure 2) in this case at 9.8 GHz. Due to the hyperfine interaction with the spin-1 nitrogens, we expect to produce six pairs of lines, with the largest splittings dictated by the dipolar electron spin-spin interaction. This is in fact observed, and enough of the transitions are resolved to permit trivial construction of a CNOT gate (in fact, irradiation on any one of the four highest frequency lines effectively does this. The dipolar splittings in this case are about 30 MHz, more than six orders of magnitude better than what would be observed with nuclear spins. In addition, while this was done with a room temperature sample at 9.8 GHz, the material is a solid, and therefore could be cooled sufficiently to achieve energy differences greater than kT (and hence scalability, which does not exist in the NMR case).

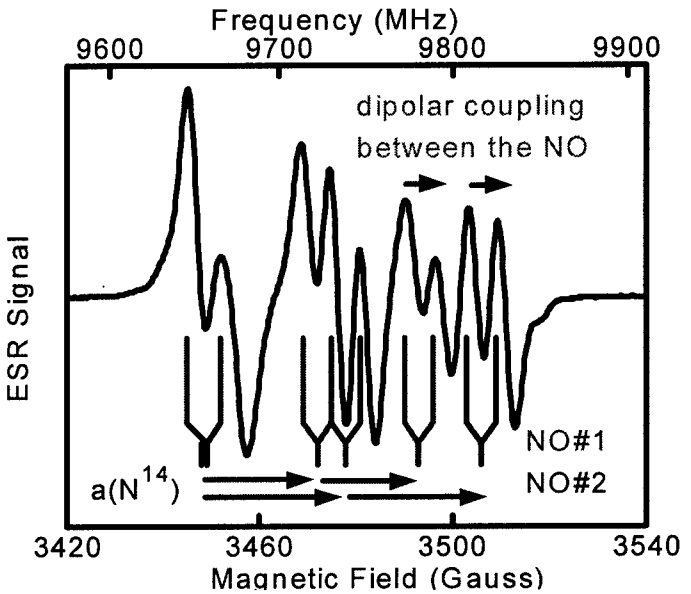

Figure 2. Experimental ESR spectrum of the molecule in Figure 1. Each nitroxide radical would have three lines (because of the spin-1 nitrogen) in the absence of coupling; the dipolar interaction generates the additional splitting.

Unfortunately, these results also point out a serious limitation of the technique. While the separation between electron spin levels could be made large enough to do cooling, the hyperfine interaction guarantees a significant nuclear spin component as well, and that implies that the ground electron spin state will be

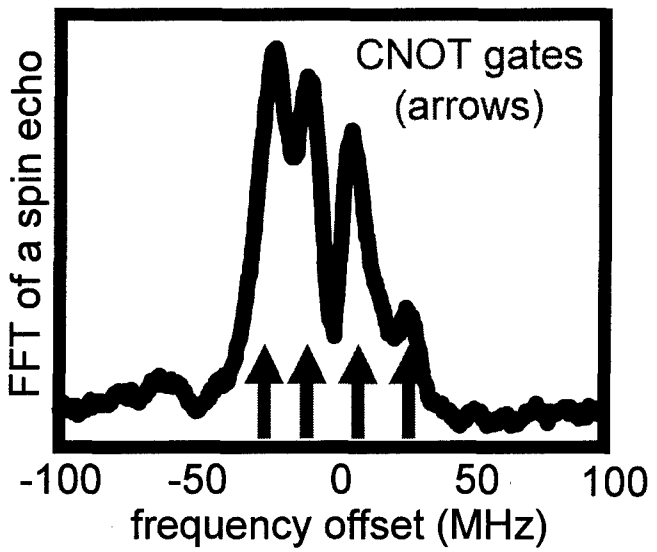

Figure 3. Pulsed spin echo ESR spectroscopy of the radical in Figure 1, showing the individual transitions that would correspond to the four lines of an effective two-qubit system. The splitting of approximately 20 MHz implies that the logic gates can be executed about 10^6 times faster than in NMR.

split into levels which cannot be cooled away. In addition, in order to really use this well as even a two-bit quantum computer, the microwave pulses would have to span at least the top four lines (ca. 100 MHz) and would have to be giving $\pi/2$ or π rotations on those lines- which is just barely within reach of the best commercial instruments. We pushed these limits in doing spin echo spectroscopy on this system at 9.8 GHz (Figure 3), and thus demonstrated the reasonably accessible resolution.

In order for this to be scalable, the excitation bandwidth (and hence the microwave

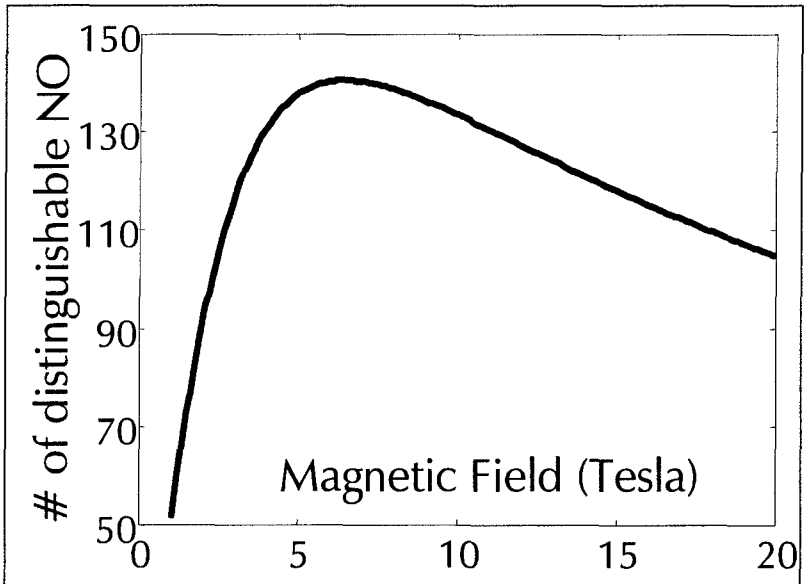

Figure 4. Theoretical calculation showing the number of resolvable transitions in principle from a favorably designed ESR system (nitroxide radicals in a glassy matrix) as a function of field strength.

power) would have to be dramatically increased. We pursued some theoretical work on the ultimate limitations of this approach, focusing on the nitroxide g-tensor anisotropy, which can range from 2.0099 to 2.0022. This implies that resonances can range over ~110 MHz / Tesla. Linewidths in glassy (water/glycerol) lattices have also been measured: the experimental linewidths (in MHz) satisfy the relationship $\nu=2+25(g-2.0022)B^2$. From this relationship it is possible to determine the number of resolvable resonances as a function of field strength. To an excellent first approximation, an N-spin quantum computer that each of the 2^N energy levels be accessible by at least one resolvable transition; thus the theoretical limits here correspond to seven or eight qubits in an optimally designed system. Unfortunately, 5 Tesla ESR corresponds to approximately 125 GHz, which requires exceedingly specialized hardware; and with today's technology, pulsed power to give $\pi/2$ or π pulses is simply not available.

Thus, while it was clear that in principle an ESR architecture would work, it was decided that technical and spectroscopic issues would make scalability be a tremendous challenge.

B. Optical Phase-Coherent pulse sequence generation

One of the major reasons why NMR quantum computing was the first demonstrated method was the relative maturity of the technological base: essentially any possible useful radiofrequency pulse sequence is trivial to generate. Optical spectroscopy offers many potential advantages (much larger couplings, energy differences greater than kT even at room temperature, much greater speed) but until recently the technology did not exist to control optical fields with the same level of precision.

In 1986 we developed the first method which gave programmable subpicosecond pulse shaping, using electro-optic modulators[1]. Subsequently Weiner modified his earlier mask-based pulse shaping technique[2] with a multielement liquid crystal modulator[3]; his group and others[4,5] have further refined the technique in recent years. In addition, there have been significant efforts using holographic patterns,[6,7] time lenses[8] and deformable mirrors.[9] Recently we have demonstrated (and patented) a new technique, acousto-optic modulation inside a zero-dispersion line, which can dramatically improve capabilities for generating complex optical waveforms (including phase and amplitude modulation).[10,11,12] This approach has advantages of rapid waveform update rates (100 kHz- 1 MHz), commercially available components (suitable AOMs have been available for decades), simplicity (the phase and amplitude modulation is encoded with a single RF pulse), and extremely high resolution pulse shaping (waveform with 1300 independently adjustable amplitudes and phases are possible, even taking into account all of the nonidealities in the laser system.)

The AOM approach permits the creation of exceedingly complex, phase controlled pulse sequences. For example, consider the apparatus as shown in Figure 5, with two radiofrequency driving waves each at a single frequency. If a single rf wave is applied, a replica of the input pulse is created. Two rf waves at different frequencies differ by a linear phase shift with time,

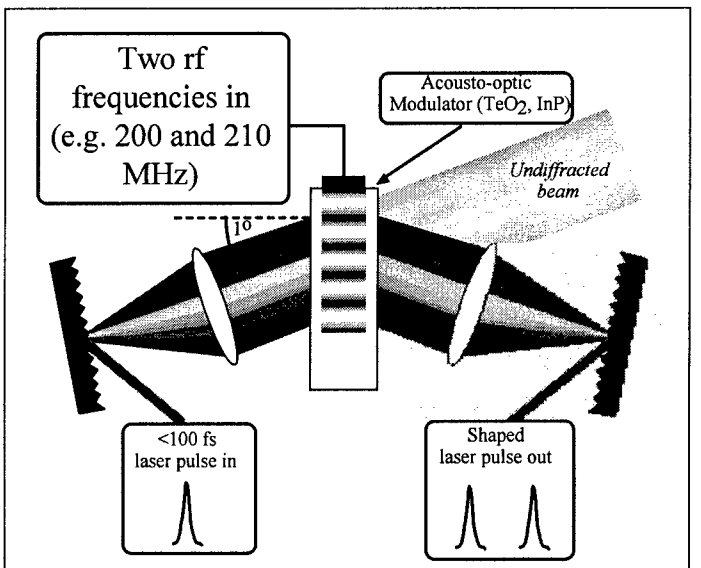

Figure 5. Acousto-optic pulse shaping method, which we have used to generate waveforms as complex as 400 phase coherent pulses.

which in turn becomes a linear phase shift with position in the modulator, and that in turn becomes a linear phase shift with frequency for the light- which is a time delay. Hence the two rf waves create two laser pulses (typically the separation is about 400 fs for a 1 MHz frequency shift), with the relative phases of the pulses dictated by the interference at the center of the modulator. Similarly, N rf waves produce an N pulse sequence.

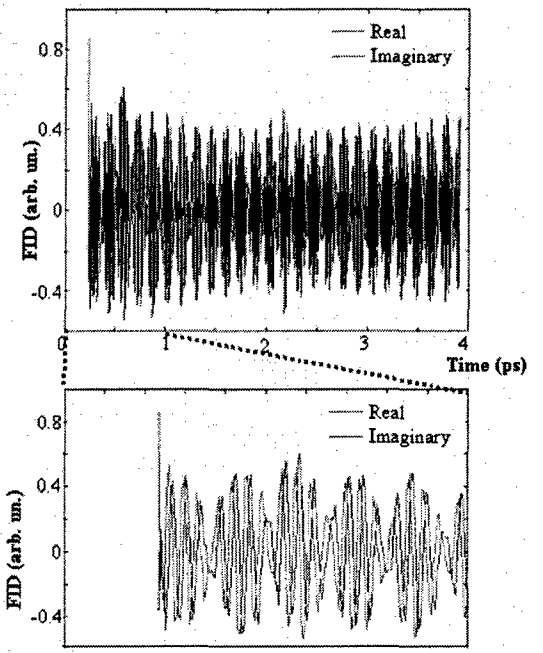

Figure 6. Experimentally measured optical free induction decay of rubidium, showing the oscillations expected by phase coherent detection.

We used this method to produce sequences with as many as 400 phase coherent optical pulses (Figure 6) and demonstrated their uses in doing phase sensitive detection of optical free induction decays. In the figure, the two optical transitions of rubidium beat against one another, and the actual phase of the oscillating optical polarization can be detected.

C. Theoretical work on quantum computing architectures

Decoherence from "Classical" Light Fields

Essentially all of the work on "bulk" quantum computing architectures (as opposed to, for example, cavity quantum electrodynamics) makes the implicit assumption that the driving fields can be treated classically. Such an assumption is perfectly reasonable for the small test systems demonstrated to date. However, any practical implementation is certain to be signal strength limited, and will work in the "efficient" regime where the net number of photons absorbed is comparable

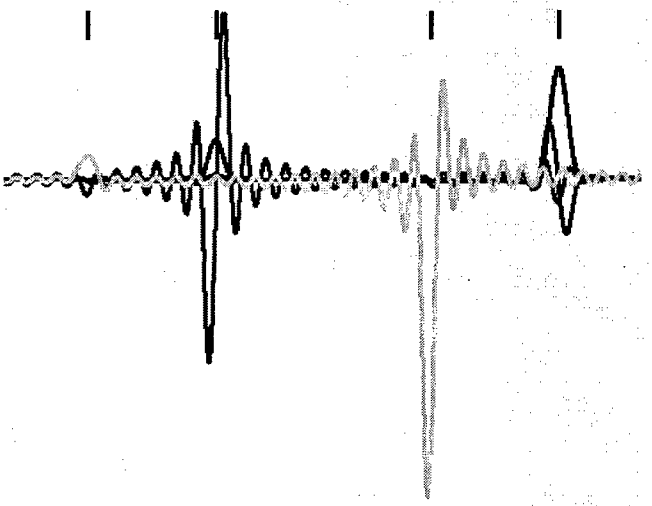

Figure 7. Decoherence in driven and undriven CNOT gates, showing additional structure added to the idealized frequency. From reference [13]

to the number of logic gates. In that limit, the statistics of the radiation field grossly perturb the logic gates;[13] in effect, the fields measure the qubits, thus collapsing the superposition states. We showed that this fundamental limit, in most idealized architectures, restricts the number of coupled qubits to 30-50: well above the limit of modern implementations, but below the point where these systems would be useful.

Automatic quantum error correction

Error correction is a fundamental and important issue for implementation of a quantum computation architecture. Unfortunately, the error correcting codes become much more complex as the number of bits increases.

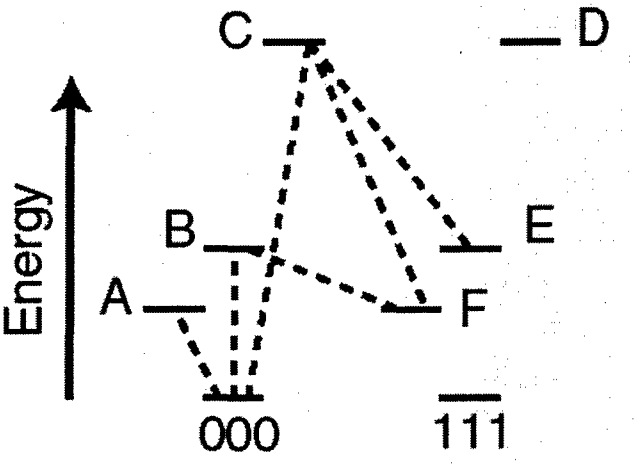

Figure 8. Energy level structure corresponding to an efficient automatic quantum error correction scheme. From reference [14].

Error correction on a classical computer is done by converting analog signals to digital values: the values "0" and "1" correspond to specific ranges of voltages,

and any voltage within that range is ultimately truncated to the optimum value. We developed an analogous scheme for a quantum computer, based on energy level diagrams with a "funnel" structure as illustrated in Figure 8. In this architecture, errors take the 0, 1 amplitude+coherence into disjoint funnel subspaces, and coupling to a cold bath returns the qubit in each funnel back to the original state. We showed that this could provide a more efficient error correcting code without as severe underlying assumptions about the nature of likely errors as is used in more conventional schemes.[14]

III. General Conclusions

The work in this proposal dramatically extended the range of applicability of fully coherent optical techniques, and was the precursor to developments of (for example) collinear two-dimensional optical spectroscopy and sophisticated pulse shaping methods for looking though tissue. However, we came to the conclusion that the technical obstacles associated with any realistically large implementation of bulk quantum computing in any molecular system are essentially unsolvable.

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