

Measurements of T_2 of electron spins at bound donor sites in Si:P

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The main activities of this program were to measure the homogeneous spin-phase memory time T_2 of electron spins on donor sites at low temperatures in Si:P and to execute a major upgrade of the ESR spectrometer to enable more comprehensive measurements. Most of the measurements were made using the pulsed ESR spectrometer in Prof. Holczer's laboratory; these were complemented by measurements carried out at Bruker Biospin Corporation. The findings include a more precise identification of the form of the echo decay than previously possible, we demonstrated the capability to make pulsed ESR T_2 measurements in Si:P at lower donor concentrations than previously reported, and we demonstrated the use of more sophisticated pulsed ENDOR techniques to probe the detailed physical origins and corresponding limits of T_2 for bound donor states in Si:P. We also demonstrated a method to measure the very long spin echo decay times of the ^{31}P nuclei at the donor sites.

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1 Introduction

The goal of the project was to establish the limitations on the transverse spin relaxation of bound donors in dilute semiconductors, in particular in Si:P. The experiments were undertaken in two steps, first using the existing pulsed EPR spectrometer in Prof. Holczer's laboratory, and then extending to longer times after obtaining an upgrade in hardware from Bruker Biospin Corporation. Below we describe the results of these activities.

2 Spin echo T_2 measurements of bound donor electrons in Si:P

As a consequence of the sensitivity of our spectrometer, we concentrated on samples with less P dopant than previously studied to confirm the low temperature concentration independence of T_2 . These results agree with the those published earlier by Chiba and Hirai [1]. Although our measurements were at the higher temperature of 8 K, the observed T_2 values are already in the temperature-independent regime observed at low temperatures. Our measurements also extended more than an order of magnitude the concentration range where the constant T_2 is observed, which makes this result very robust. A similar saturation of T_2 is observed as a function of temperature; around 10-12K. Above this temperature, $T_1=T_2$; below it, the observed T_1 and T_2 values separate as the echo decay develops a strongly non-exponential shape. This behavior was observed previously, and was interpreted as a consequence of nuclear spin diffusion involving mutual spin flips of ^{29}Si nuclei. For comparison to Ref. [1], we include the data of Fig. 1.

The shape of the echo decay appears to stabilize at the same temperature at which it becomes temperature independent. In contrast, T_1 continues to increase exponentially. The exact shape of the low temperature phase-memory decay is consistent with the following empirical form,

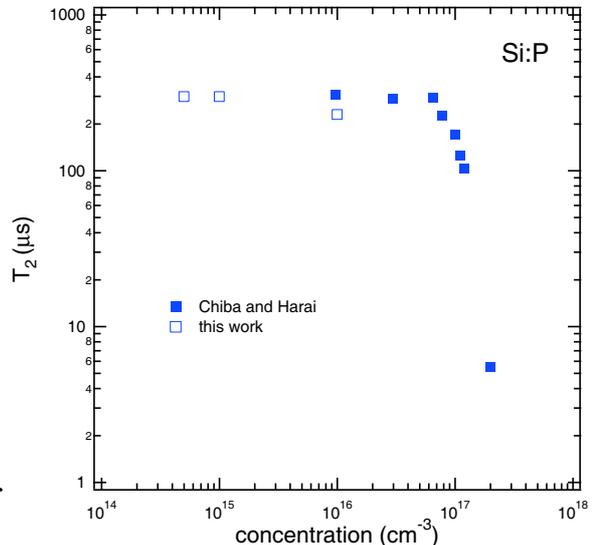


Figure 1: T_2 versus Phosphorous concentration at $T=1.6\text{K}$ (filled squares, M.Chiba, and A. Hirai, Ref. [1]). The open circles show the three samples studied by us at $T=8\text{K}$.

$$S(2\tau) = S_0 e^{-(2\tau/T_2)^\alpha}, \quad (1)$$

with $\alpha = 9/4$.

The evolution of the shape from the Lorentzian form at higher temperatures, to the form described in Eq. [1] is very clear from the data shown in Fig. 2. Figure 3 shows that within the experimental uncertainties, our measurement very similar to the earlier results of Ref. [1]. However, our results sufficiently precise to demonstrate that the decay differs in a significant way from their proposed analytical form. Still, we cannot exclude an eventual (small) change in shape at lower temperatures, nor have we examined systematically the effect of microwave power, sample orientation, etc. on the decay shape.

The Arrhenius plot of Fig. 4 shows the temperature dependence of T_1 and T_2 together. The rate T_1 is dominated by the thermally excited electrons in the conduction band. From the curve, one obtains the value $\Delta E=15\text{meV}$ for the activation energy of the bound donor states. Above $T \approx 30\text{-}35\text{K}$, no EPR signal is observed,

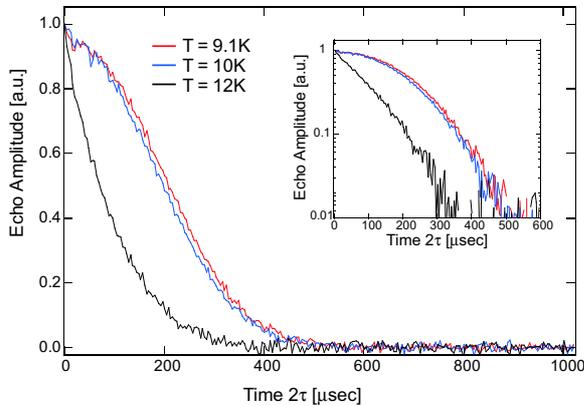


Figure 2: Comparison of the normalized Phase Memory (Hahn Echo) decay curves at three temperatures. Below 12 K, a single, non-exponential form is observed. The insert shows a semi-log plot of the same data.

in part because the sample conductivity is high enough to create substantial microwave losses. Also, both T_1 and T_2 are too short to detect a spin echo signal. It is interesting to point out that the EPR signal is inhomogeneously broadened over the whole range of temperature. This width, $\Delta H \sim 2G$, makes it possible to generate the echo signal. At the higher temperatures, T_2 has the same value and is determined by T_1 . They have the same value down to about $T=10$ - $12K$, below which T_2 levels off and T_1 continues its activated increase. The mechanism responsible for the low temperature, temperature-independent, part of T_2 are the local field fluctuations caused by ^{29}Si nuclear spin diffusion. These result from energy-conserving mutual ^{29}Si spin flips.

This temperature dependence of T_2 is a novel, although not unexpected behavior. It is worth noting that the value of $250\mu s$ found for the T_2 of donors in lightly-doped Si:P is by most standards quite long. A selected list of such "long" T_2 materials is shown in Table 1. Note that all of the other values listed are at room temperature and one of them is in solution. These are, however, samples commonly regarded as hav-

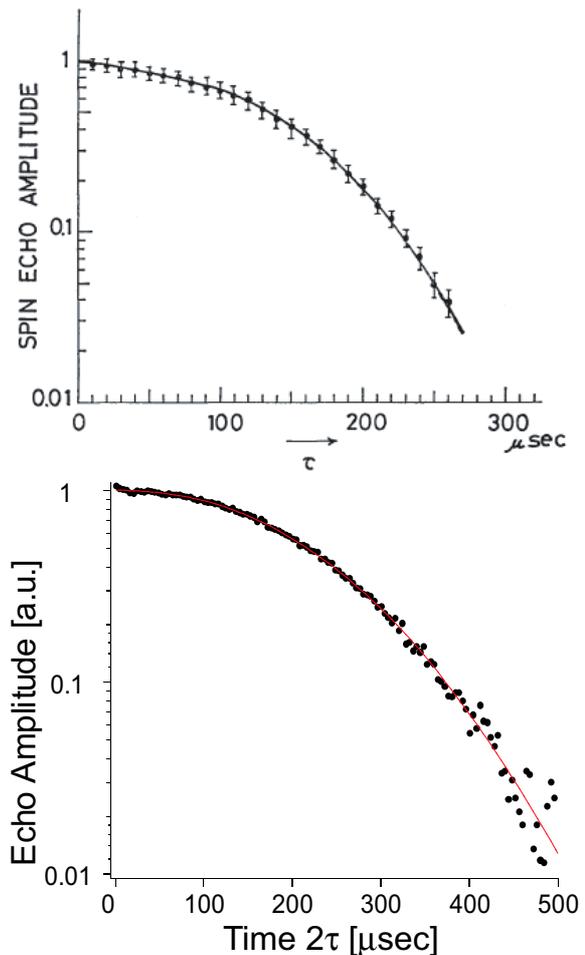


Figure 3: A comparison of our measurements of the spin echo decay shape with those of Ref. [1] (top). The functional forms of the fits are $S(\tau) = \exp -[(\tau/T_2)^3 + \tau/T_1]$ (top, Ref. [1]), and Eq. [1] (bottom)

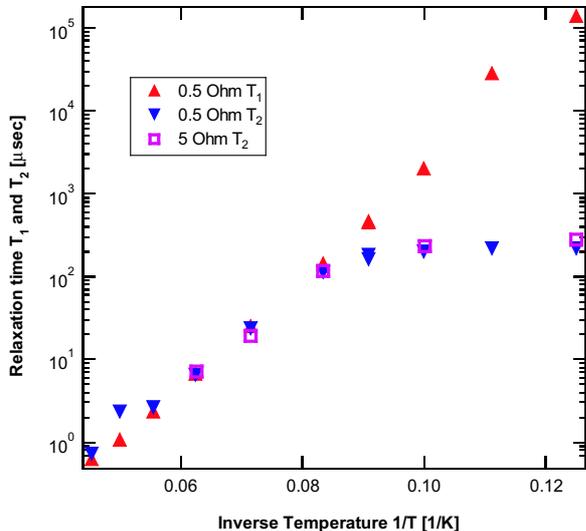


Figure 4: Temperature dependence of T_1 and T_2 of the three samples investigated.

ing extremely long values for the spin echo decays. The non-exponential decay and time scale for echo decay of the Si:P system are not intrinsic, in the sense that it is not controlled directly by the state of the bound donor and the temperature. It arises from random local field fluctuations associated with the distant ^{29}Si nuclear spins. Their effectiveness in suppressing the electron spin echo can be removed with appropriate pulse sequencing, such as variations on the Carr-Purcell sequence [2], by removing the low-abundance ^{29}Si nuclei from the system, or by removing their effect through RF decoupling techniques. The nuclear spins causing the relaxation are distant; only these can undergo energy-conserving mutual spin flips. Closer nuclear spins are in sufficiently different local fields so as to suppress spin flips.

3 Pulsed electron-nuclear double resonance (ENDOR) measurements

Pulsed Electron-Nuclear Double Resonance (ENDOR) experiments provide a sophisticated way to explore the properties of a many-spin system such as this. In other words, the electron

material	conditions	T_2
γ irradiated quartz	300K	2-5 μs
N centers in diamond	300K	2-100 μs
N in N@C ₆₀	in solution	120 μs
P in Si:P	T<12K	250 μs

Table 1: A short list of long T_2 values in comparison with the value found in n-type Si.

spin strongly coupled to the $I=1/2$ spins of the ^{31}P (100% natural abundance) and the randomly distributed ^{29}Si isotopes (4.67% natural abundance). Pulsed ENDOR allows going one step further to perform nuclear spectroscopy through EPR detection. While this technique allows one to acquire the spectrum of each coupled nuclear spin, it is not limited to it. The response to more complex NMR excitations are equally accessible within the time limit set by the electronic T_1 . The latter is an exponential function of the temperature and can therefore be controlled. With this method, we can also measure T_2 for the nuclear spins.

In Fig. 5, we show the ^{29}Si ENDOR result, which demonstrates the strong coupling of the far-away nuclear spins to the electron spin. The RF frequency is varied while simultaneously detecting the electron spin echo (ESE). As the RF passes the Larmor frequency of the distant ^{29}Si spins, the echo amplitude is suppressed strongly. With higher amplitude RF, the echo should be restored through motional-narrowing effects. In other words, the long spin phase memory time can be restored by decoupling the nuclear spins.

The possibility to use the dopant site as a double qubit, *i.e.*, utilize both the electron and the nuclear spin in a future quantum computer, has been suggested [3]. In this application, both the electron and the nuclear spin decoherence times are crucial. To the best of our knowledge, the pulse sequence shown in Fig. 6 is the only possibility for measuring the macroscopic T_2 of the relevant nuclei. In Fig. 7, we show the ^{31}P pulsed ENDOR signal at T=8K, which exhibited a ^{31}P nuclear T_2 equal to the electron T_1 .

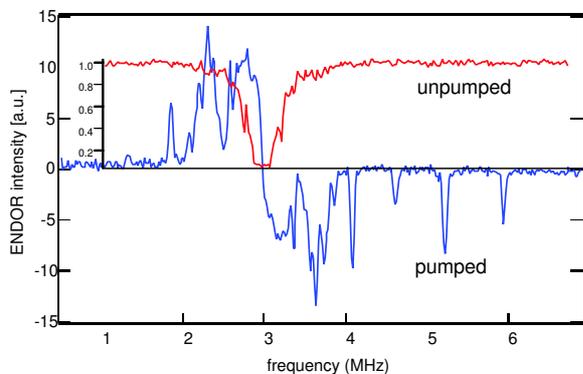


Figure 5: ^{29}Si ENDOR spectra. The red curve demonstrates the ESE suppression when the RF matches the Larmor frequency of the distant nuclei.

References

- [1] M. Chiba and A. Hirai, J. Phys. Soc. Japan **33**, 730 (1972).
- [2] A. M. Tyryshkin, *et al.*, Phys. Rev. B **68**, 193207 (2003).
- [3] B. E. Kane, Nature **393** 133 (1998).

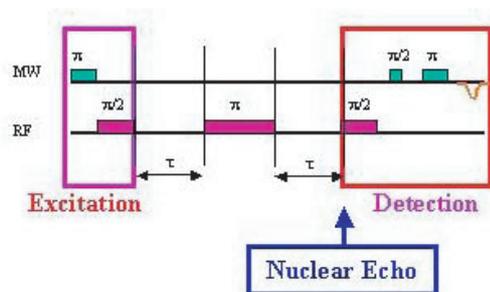


Figure 6: ENDOR pulse sequence used to detect the nuclear phase memory time T_2 . The third RF pulse is applied at the expected NMR center, as the EPR detection sequence is sensitive to the z-component of the nuclear magnetization. The EPR excitation/detection sequence duration is limited to T_1 of the electron.

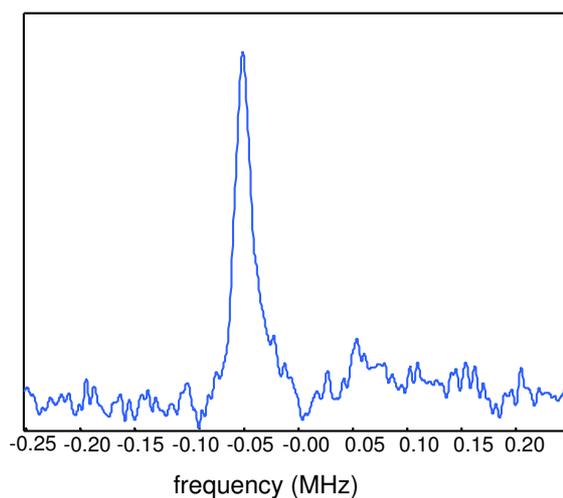


Figure 7: ^{31}P pulsed ENDOR signal. The method uses a pulse sequence where the electron spin echo amplitude is sensitive to the z-component of the nuclear magnetization.