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Physical nature of electrically detected magnetic resonance through spin dependent trap assisted tunneling in insulators

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We show that electrically detected magnetic resonance (EDMR), through spin dependent trap assisted tunneling (SDTT) in amorphous SiC, exhibits approximately equal amplitudes at very high (8.5 T) and very low (0.013 T) magnetic fields at room temperature. This result strongly supports an SDTT/EDMR model in which spins at two nearby sites involved in a tunneling event are coupled for a finite time in circumstances somewhat analogous to spin pair coupling in the spin dependent recombination/EDMR model of Kaplan, Solomon, and Mott (KSM) [Kaplan, Solomon, and Mott, J. Phys. Lett. **39**, 51 (1978)]. Since a comparable near zero magnetic field change in resistance is also observed in these samples, our results support the idea that this magnetoresistance response is also the result of a KSM-like mechanism involving SDTT. Additionally, we observe a large enhancement in SDTT/EDMR at high field (8.5 T) for temperatures below 50 K, which suggests the potential utility of SDTT in spin based quantum computation and other spintronic applications. *Published by AIP Publishing.* https://doi.org/10.1063/1.5057354

I. INTRODUCTION

Electrically detected magnetic resonance (EDMR) is a topic of great interest,[1,2] in part for its obvious relevance to quantum computation,[3,4] but also for the opportunity that it provides for understanding electronic transport mechanisms in semiconductors and insulators.[5–7] EDMR involves quite sensitive detection of electron paramagnetic resonance (EPR) through changes in the current or voltage measured in a device structure simultaneously exposed to both an oscillating and quasi-static magnetic field. The EDMR detection of spin dependent recombination (SDR) has been extensively utilized in numerous studies. Its underlying physical mechanisms are, to a large extent, understood in terms of models which evolved from a seminal paper by Kaplan, Solomon, and Mott (KSM).[1]

A second spin dependent mechanism which can be detected through EDMR is spin dependent trap-assisted tunneling (SDTT). SDTT, enabled through variable range hopping, exploits the fact that spin angular momentum is conserved in trap to trap tunneling events as qualitatively illustrated in Fig. 1. Like SDR/EDMR, SDTT/EDMR has the potential for quantum computation and electronic materials physics. The limited SDTT/EDMR literature on the latter[6–8] presents very little direct evidence with regard to the detailed physical mechanisms involved. In this paper, we provide physical insight into the SDTT/EDMR process as it occurs in a technologically important material,[9,10] amorphous hydrogenated SiC (a-SiC).[8,11] We provide evidence that the SDTT/EDMR mechanism in this system is, in a fundamental respect, analogous to the KSM process. We also show that a very large enhancement in the SDTT/EDMR response can be achieved with low temperature and high magnetic fields. In addition, we note that a substantial change in resistance in these films occurs at and near zero magnetic field. As reported previously, the magnitude of this near zero field change is roughly comparable to the change induced by SDTT/EDMR.[8] Thus, our results also provide a strong indication that the low field magnetoresistance effect in these materials is also due to a KSM-like mechanism involving SDTT.

Our observations may be of interest for materials other than a-SiC. For example, amorphous silicon, a-Si, exhibits an EPR response which is strongly correlated with the density of states within the bandgap.[12] The response is due to silicon dangling bond centers in the a-Si.[13] This EPR response can be greatly reduced by the presence of a high concentration of hydrogen within the amorphous silicon.[12] The leakage current density observed in a-SiC is strongly correlated with the magnitude of the EPR response.[11] The paramagnetism and leakage currents are also strongly correlated, strongly suggesting a correspondence between the resonance and bandgap states in the a-SiC.[11] In addition, the ESR response in a-SiC may also be greatly reduced by a high concentration of hydrogen.[11] Furthermore, electronic transport in a-Si has been associated with variable range hopping,[12,14,15] which we argue is the likely dominating transport mechanism in these a-SiC structures.

The seminal KSM paper[1] explained SDR/EDMR in terms of the behavior of a pair of spins involved in recombination, a pair now generally acknowledged to be spin associated with a deep level defect and spin associated with a charge carrier. A surprising aspect of the KSM model is its prediction that the SDR response is, to the first order, independent of the field and frequency at which the measurement is made. KSM explained that this remarkable field and frequency response arises because a spin dependent recombination event does not involve an instantaneous collision-like

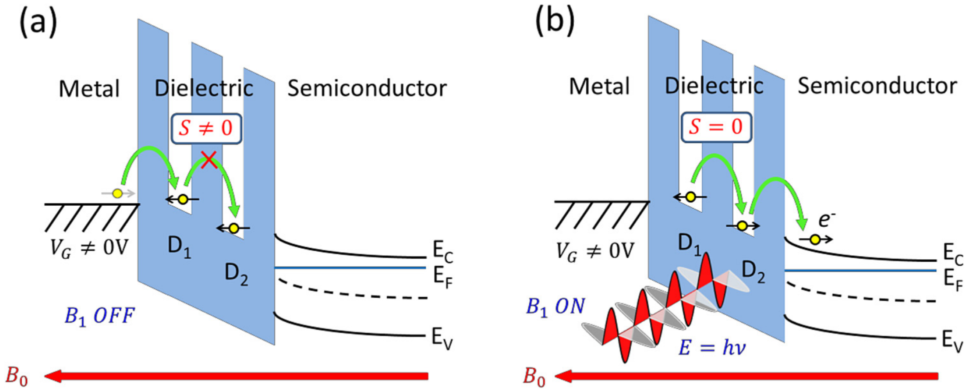

FIG. 1. A generalized illustration of spin dependent trap assisted tunneling involving two paramagnetic defect sites. Tunneling would be allowed from one defect to the other if spin angular momentum is conserved; this would be the case for a singlet pair. Tunneling from one defect to the other is forbidden if angular momentum is not conserved, the case for a triplet pair. However, if electromagnetic radiation satisfying the resonance condition is present, that radiation can "flip" paramagnetic defect spins, rendering previously forbidden tunneling events allowed, thereby increasing current across the dielectric.

event, but a process involving the coupling of two spins for a finite amount of time. In the KSM SDR model, a pair of electron spins, initially in a singlet state, will immediately take part in a capture event, whereas a pair of spins initially in a triplet state may take part in a recombination event only at a greatly decreased rate which will depend upon electron spin relaxation times, a rate which can be greatly increased by magnetic resonance induced transitions. (Essentially, the triplet must be transformed to a singlet for the process to take place.) Thus, for relatively high temperatures and small to very large fields, the KSM response is, to first order, independent of electronic polarization, and thus independent of the field and frequency at which the SDR/EDMR measurement takes place.

One might envision two general SDTT/EDMR response categories: one in which a pair of spins is involved in a tunneling process which takes a finite time, as in the KSM SDR model, or an instantaneous collision-like process analogous to an early SDR/EDMR model developed by Lepine.[2] If the SDTT/EDMR process involves an essentially instantaneous collision-like process, as Lepine showed, the magnitude of the effect would scale with the square of the electron polarization and thus the square of the field of resonance. This result grossly contrasts with a coupled spin-pair like model in which, as KSM proposed and others have demonstrated,[16,17] there is a near field independent response. However, at sufficiently low temperatures and high fields, the difference in electron polarization yields such a large difference between singlet and triplet configurations that the response would inevitably grow significantly larger. (Longer spin-spin relaxation times at a lower temperature could also contribute to the very large high field low temperature response.) We perform our measurements at magnetic fields of approximately 0.013 and 8.5 T, fields which vary by over a factor of 650. If the SDTT/EDMR process is Lepine-like, one would anticipate an enormous difference in the two responses. For the case of measurements made at room temperature, the difference would be more than a factor of 4×10^5. In contrast, if the SDTT/EDMR process is KSM-like, one would anticipate approximately equal responses at each field.

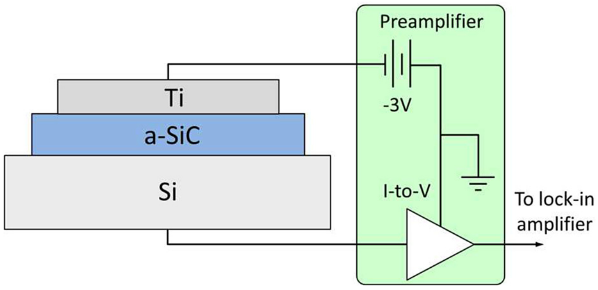

FIG. 2. Illustration of the biasing and current monitoring in our experimental setup.

II. EXPERIMENTAL

We have conducted EDMR measurements on 5 nm films of a-SiC deposited on (100) p-type silicon substrates with titanium caps. In all measurements, a potential of −3 V was applied to the Ti caps. The sample biasing arrangement is schematically illustrated in Fig. 2. The low (0.013 T) magnetic field EDMR measurements were made at Penn State using a home built instrument in which the oscillating magnetic field was provided by a surface coil in a simple inductor-capacitor tuned circuit which was impedance matched to an RF signal generator (Stanford Research Instruments Model SG382). The low magnetic field was generated by a home built electromagnet consisting of five Helmholtz coils with field control provided by a computer, a temperature compensated Hall probe with Lake Shore 450 Gaussmeter, and a Kepco power supply. We amplitude modulated the quasi-static magnetic field at audio frequencies and demodulated using a virtual lock-in amplifier. As a result, the low-field spectra are reported herein as first derivatives. The high (8.5 T) magnetic field measurements were made at the National High Magnetic Field Laboratory (NHMFL) using one of the quasi-optical spectrometers.[18] These spectrometers utilize modulation of the oscillating electromagnetic field amplitude and therefore produce spectra that represent the SDTT absorption, rather than its derivative. For both spectrometers, we utilize the Stanford Instruments SR570 preamplifier for biasing the dielectric, amplifying device currents, and analog signal conditioning prior to demodulation. Although it is difficult or impossible to provide precisely equivalent circumstances for both measurements, the oscillating magnetic field amplitudes, B_1, were approximately matched at 0.15 Gauss. It is important to note that, although this oscillating field amplitude generates a strong response, the response is far from saturated in all measurements. However, as pointed out by Kawachi *et al.*,[19] one would expect the EDMR amplitude to grow approximately with the square root of RF or microwave power when the system is weakly saturated and, at a very high power, the amplitude would be constant. Although we were not able to achieve full saturation in either the low or high field and frequency case, in both cases, the measurements were in the weakly saturated regime.

III. RESULTS

Figure 3 illustrates the SDTT/EDMR results obtained on the a-SiC dielectric at high magnetic fields for various temperatures. Panel (a) illustrates a representative SDTT/EDMR spectra acquired with $T = 300$ K, and panel (b) illustrates the peak amplitude of the SDTT/EDMR response and the DC leakage current versus temperature. (The reasons for the U shaped ΔI vs temperature behavior are presented in Sec. IV.) One might note that the current versus temperature results in panel (b) cannot be fit to the classical $\sigma = A\ \exp(-B/T^{1/4})$ model first proposed by Mott.[12] The classical Mott expression is however plausibly consistent with our results at lower temperatures. As illustrated in panel (c), the natural logarithm of current plotted versus $T^{-1/4}$ is linear at lower temperatures. As has been discussed by others previously, the $\exp(-B/T^{1/4})$ behavior does not accurately describe transport in a-Si above about 200 K.[14,15] Mott's result, although quite useful, is based on a significantly simplified model. More sophisticated models, such as that of Efros and Shklovskii,[20] for example, predict a somewhat different behavior.

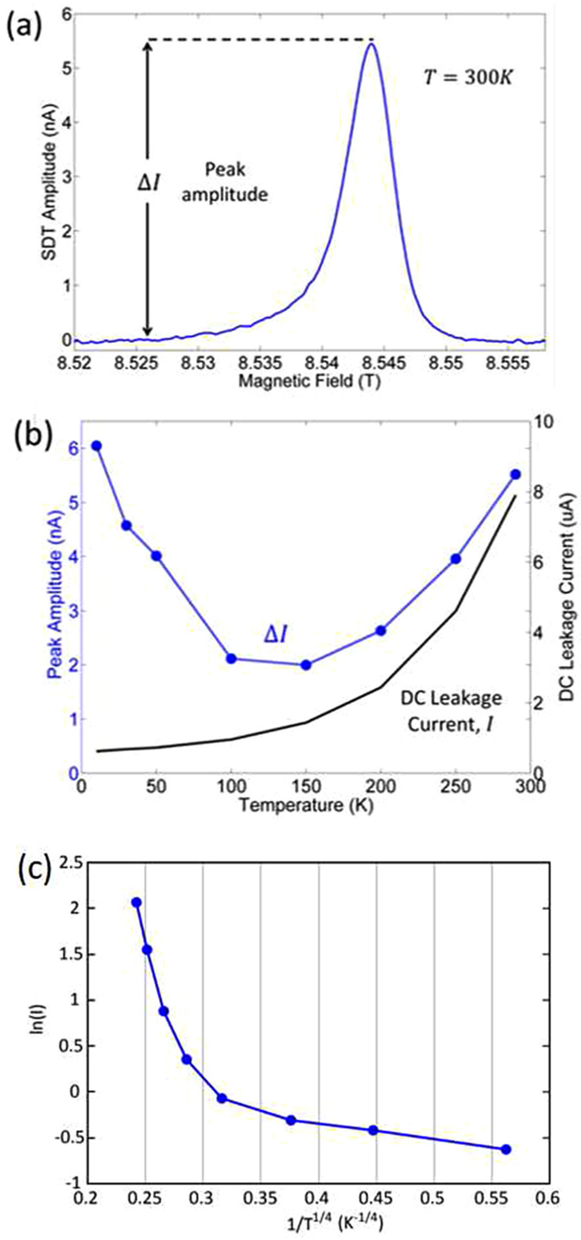

FIG. 3. Illustration of (a) a representative high-field SDTT/EDMR trace acquired at 300 K, the g corresponding to the peak is 2.0030 ± 0.0002; (b) peak amplitude of this response and DC leakage current versus temperature; and (c) DC leakage current vs $T^{-1/4}$ which is linear at low temperatures.

Figure 4 illustrates additional SDTT/EDMR data, all obtained on the a-SiC dielectrics at very low to zero magnetic fields. Panel (a) illustrates low-field measurements taken at 300 K, 250 K, 200 K, and 150 K. The low-field measurements were made with a modulation amplitude (≈0.3 mT) significantly less than the width of resonant response (≈0.8 mT) which enabled us to integrate the data as a reasonable approximation of the first derivative, thereby allowing a meaningful $A\ \exp(-B/T^{1/4})$ comparison with the high field absorption data which is illustrated in Fig. 5. A brief explanation of our integration process may be useful. The derivative plots of Fig. 4 represent the output voltage from a lock-in amplifier; the lock-in input is the output of a current to voltage amplifier which monitors the spin dependent current from the a-SiC sample. Taking into account the conversion factor of the current to voltage amplifier and the lock-in amplifier gain, the lock-in output is calibrated to yield an approximate derivative, di/dB, where i is the device current and B is the (0.3 mT) modulation field amplitude. [The amplitudes involved in the plots of Fig. 4(a) correspond to the change in device current with 0.3 mT modulation amplitude.] Integrating the lock-in output thus yields a moderately accurate integral suitable for comparison with the high field data which was measured utilizing microwave amplitude modulation. As illustrated in the figure, two different responses are present: the low-field (0.013 T) SDTT/EDMR and the zero-field response (magnetoresistance). Panel (b) illustrates the corresponding peak amplitudes (peak-to-peak derivative current response) of both the zero-field and low-field responses for the spectra illustrated in (a) plotted against the measured DC leakage as a function of temperature from room temperatures to 150 K. Although the low field measurements were made over a fairly limited temperature range, over this range the low field SDTT/EDMR response is qualitatively similar to that observed at high fields. We believe these low-/zero-field measurements may be of widespread interest. Recently, relatively low field magnetoresistance has been reported in organic semiconductors, tunneling in double quantum dots and in a-SiC, as well as SDR dominated transport in SiC diodes.[5,8,21–26]

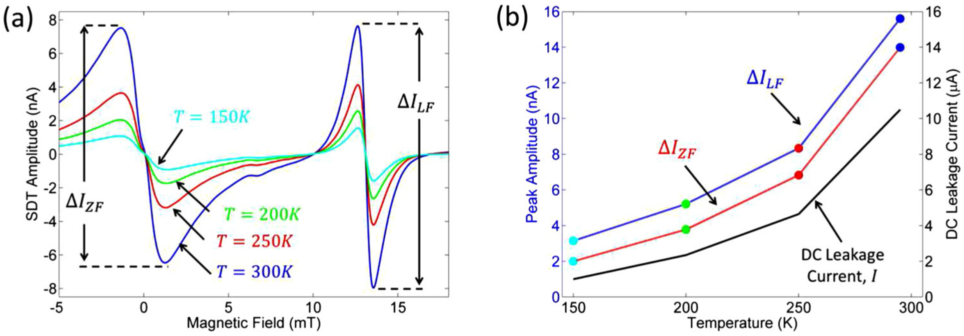

FIG. 4. Illustration of representative (a) zero- and low-field (0.013T) spectra and (b) their corresponding peak amplitudes for various temperatures plotted along with the measured DC leakage current. For the temperatures utilized in this illustration, the SDTT amplitude and qualitative temperature response are similar to the response at high field.

Figure 5 summarizes a key result from the high- and low-field measurements: roughly comparable room temperature amplitudes for high-field spectrum and integrated low-field spectrum. Recall that a Lepine-like process would be expected to yield about a field squared ratio in EDMR amplitude. This would correspond to about $(8.5/0.013)^2 \approx 430\,000$. A KSM-like response would be expected to be roughly field independent. Note again that these spectra were obtained using identical biasing conditions ($V = -3$ V), amplifier settings, and approximately the same oscillating magnetic field of $B_1 \approx 0.15G$. Figure 6 compares the magnetic resonance change in current divided by the DC leakage current ($\Delta I/I$) for the high-field (8.5 T) response versus temperature. Note the quite large increase in $\Delta I/I$ at the lowest temperatures.

IV. DISCUSSION

Of particular interest is the comparison of the high and low field responses in Fig. 5. These spectra correspond to a zero crossing g of 2.0030 ± 0.0002 and are likely due to a dangling bond center in the amorphous SiC.[8] The SDTT/EDMR amplitudes at the two frequencies are within about a factor of three of one another, with the low field response about three times as high as the high-field response. Since a KSM-like physical mechanism would yield roughly equal responses and a Lepine-like physical mechanism would yield a reduction in low field response of about a factor of 4×10^5, all things being equal, this simple result leads to a very straightforward conclusion. Since our result differs from the prediction of a Lepine-like collision mechanism by a factor of about 12×10^5 but is consistent with an anticipated approximately equal KSM-like finite time pairing response, we conclude that the SDTT process involves a finite time coupling between two spins at nearby paramagnetic sites taking part in variable range hopping transport.

Also of particular interest is a plot of the high-field $\Delta I/I$ versus temperature illustrated in Fig. 6. One would expect to see a large increase in the SDTT/EDMR response at a lower temperature if the ratio of magnetic field to temperature were sufficiently high so as to create a polarization in which the difference between the two possible spin orientations becomes large enough to overcome the KSM mechanism response. A simple calculation allows for an order-of-magnitude estimate of the circumstances required for a spin-polarization dominated response. The polarization P of a system of nearly isolated electron spins (spin ½ sites for which $g = 2$, a good approximation in our case) is given by $P(T) = \tanh(\mu_B B_0/kT)$.[27] Here, μ_B is the Bohr magneton, B_0 is the magnetic field at which the measurement is made, k is the Boltzmann constant, and T is the absolute temperature. One would expect to observe a Lepine-like response should the product of the two spin polarizations exceed that of the ratio involved in the KSM-like

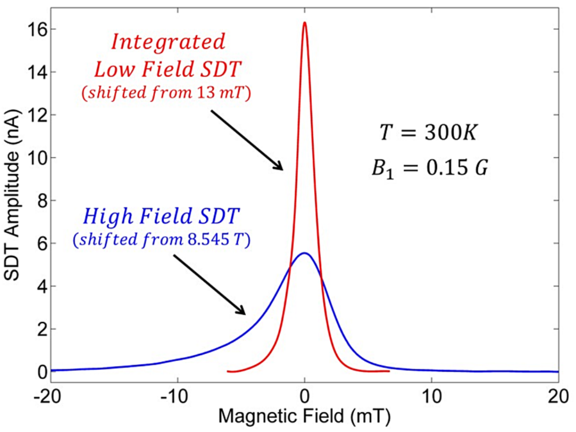

FIG. 5. Comparison of the high-field (8.545 T) and integrated low-field (0.013 T) responses at room temperature. Biasing conditions were identical in both cases. The similarity in the amplitude of the responses is sufficiently close which indicates that the EDMR mechanism involves a finite coupling time spin pair response.

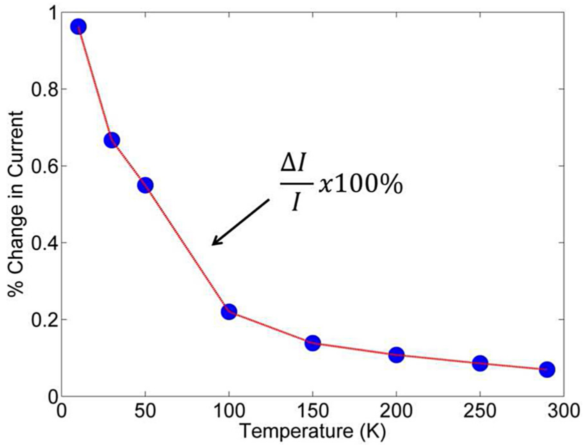

FIG. 6. An illustration of the high field SDTT response as a function of temperature. At room temperature, the response is about 0.07%. The measured response at lower temperatures is quite large, reaching about 1% at 10 K. At lower temperatures and higher $B1$, it is virtually certain that a much larger response could easily be achieved.

behavior. Since the magnitude of the KSM response itself depends to some extent on the temperature, it is difficult, if not impossible, to precisely quantify such a transition. Nevertheless, at a field of 8.5 T, the ratio $\mu_B B_0/k = 5.7$; so the polarization reaches about 10% at about 57 K and about 52% at 10 K. Since the room temperature SDTT/EDMR response corresponds to a change of about 0.07%, one would anticipate the possibility of a transition from KSM to spin polarization dominated behavior, and a much larger SDTT/EDMR response at even moderately low temperature and such a high field which is clearly illustrated in Fig. 6. The results are unfortunately limited to only a moderately low temperature (10 K) because the a-SiC measurements involved a silicon substrate/a-SiC dielectric/metal sandwich structure. The high resistance of the dielectric totally dominates the response at higher temperatures but, at extremely low temperatures, the silicon substrate resistivity would eventually confound the results.[28,29] Thus, we limited our lowest temperature measurements to 10 K. Nevertheless, the qualitative response is clear: at low temperatures, there is a very large total increase in the ratio of SDTT current change with respect to current. At the lowest temperature, 10 K, the response is about 1%. This large effect could almost certainly be substantially increased to increasing the B_1 magnetic field and or even a modest additional decrease in the measurement temperature. (It should be noted that although a shift in SDTT response from a KSM to a Lepine like model is a reasonable interpretation of this low temperature response, it is not absolutely conclusive since a KSM like response also depends upon other temperature dependent factors, among them spin lattice relaxation time.)

V. CONCLUSION

Our results, most importantly, provide very strong evidence that the SDTT/EDMR mechanisms observed herein at higher temperatures involve a finite time pair coupling process somewhat similar to that first articulated for SDR/EDMR by KSM. In this case, the coupling would involve coupling between two paramagnetic sites involved in trap to trap tunneling. Our results also demonstrate a strong enhancement in SDTT/EDMR at low temperature and high field. Although the ratio of EDMR response vs total current monotonically decreases with increasing temperature, the absolute amplitude of the EDMR is greatest at both the highest and lowest temperatures investigated. This is so because the absolute current is the highest at the highest temperature investigated, and the magnetization and spin lattice relaxation times are the highest at the lowest temperatures investigated. With the B_1 field very far from a saturation value for the SDTT, the measured response at the modest temperature of 10 K approaches 1%, indicating that much larger effects should be achievable at lower temperatures and higher values of B_1. This observation may be of considerable interest for spin based quantum computation and other spintronic applications, as it demonstrates the potential for extremely sensitive EDMR at very low temperatures, at which decoherence times are almost certainly the longest.

ACKNOWLEDGMENTS

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[1] D. Kaplan, I. Solomon, and N. F. Mott, J. Phys. Lett. **39**, 51 (1978).

[2] D. J. Lepine, Phys. Rev. B **6**, 436 (1972).

[3] I. T. Vink, K. C. Nowack, F. H. L. Koppens, J. Danon, Y. V. Nazarov, and L. M. K. Vandersypen, Nat. Phys. **5**, 764 (2009).

[4] J. J. L. Morton, D. R. McCamey, M. A. Eriksson, and S. A. Lyon, Nature **479**, 345 (2011).

[5] C. J. Cochrane, P. M. Lenahan, and A. J. Lelis, Appl. Phys. Lett. **100**, 023509 (2012).

[6] J. P. Campbell, P. M. Lenahan, A. T. Krishnan, and S. Krishnan, J. Appl. Phys. **103**, 044505 (2008).

[7] J. T. Ryan, P. M. Lenahan, A. T. Krishnan, and S. Krishnan, J. Appl. Phys. **108**, 064511 (2010).

[8] C. J. Cochrane and P. M. Lenahan, Appl. Phys. Lett. **104**, 093503 (2014).

[9] Y. Matsuda, J. S. Rathore, L. V. Interrante, R. H. Dauskardt, and G. Dubois, ACS Appl. Mater. Interfaces **4**, 2659 (2012).

[10] C.-C. Chiang, I.-H. Ko, M.-C. Chen, Z.-C. Wu, Y.-C. Lu, S.-M. Jang, and M.-S. Liang, J. Electrochem. Soc. **151**, G93 (2004).

[11] T. A. Pomorski, B. C. Bittel, C. J. Cochrane, P. M. Lenahan, J. Bielefeld, and S. W. King, J. Appl. Phys. **114**, 074501 (2013).

[12] N. F. Mott, *Conduction in Non-Crystalline Materials* (Clarendon Press, Oxford, 1987).

[13] D. K.Biegelsen and M. Stutzmann, Phys. Rev. B **33**, 3006 (1986).

[14] S. K. Bahl and S. M. Bhagat, J. Non-Cryst. Solids **17**, 409 (1975).

[15] K. Morgaki, in *Physics of Amorphous Semiconductors* (Imperial College Press, London, 1999), Chap. 7.

[16] W. Akhtar, H. Morishita, L. S. Vlasenko, D. S. Poloskin, and K. M. Itoh, Phys. B Condens. Matter **404**, 4583 (2009).

[17] M. S. Brandt, M. W. Bayerl, M. Stutzmann, and C. F. O. Graeff, J. Non-Cryst. Solids **227–230**, 343 (1998).

[18] G. W. Morley, L.-C. Brunel, and J.van Tol, Rev. Sci. Instrum. **79**, 064703 (2008).

[19] G. Kawachi, C. F. O. Graeff, M. S. Brandt, and M. Stutzmann, Jpn. J. Appl. Phys. **36**, 121 (1997).

[20] A. L. Efros and B. I. Shklovskii, J. Phys. C Solid State Phys. **8**, L49 (1975).

[21] C. J. Cochrane and P. M. Lenahan, Appl. Phys. Lett. **103**, 053506 (2013).

[22] W. J. Baker, K. Ambal, D. P. Waters, R. Baarda, H. Morishita, K. van Schooten, D. R. McCamey, J. M. Lupton, and C. Boehme, Nat. Commun. **3**, 898 (2012).

[23] Z. H. Xiong, D. Wu, Z. Valy Vardeny, and J. Shi, Nature **427**, 821 (2004).

[24] Y. Sheng, T. D. Nguyen, G. Veeraraghavan, Ö. Mermer, M. Wohlgenannt, S. Qiu, and U. Scherf, Phys. Rev. B **74**, 045213 (2006).

[25] F. H. L. Koppens, C. Buizert, K. J. Tielrooij, I. T. Vink, K. C. Nowack, T. Meunier, L. P. Kouwenhoven, and L. M. K. Vandersypen, Nature **442**, 766 (2006).

[26] N. J. Harmon and M. E. Flatté, Phys. Rev. B **85**, 075204 (2012).

[27] R. P. Feynman, R. B. Leighton, and M. Sands, *Feynman Lectures on Physics* (Addison Wesley, Reading, MA, 1964), Vol. 2, Chap. 35.

[28] J. S. Blakemore, *Semiconductor Statistics* (Dover Publications, Mineola, NY, 1987), Chap. 3.

[29] R. H. Bube, *Electronic Properties of Crystalline Solids* (Academic Press, New York, NY, 1974), Chap. 9.