NUMERICAL STUDY OF SPIN-DEPENDENT ELECTRONIC TRANSITION RATES BETWEEN TWO DIPOLAR AND EXCHANGE COUPLED PARAMAGNETIC (S=1/2) STATES DURING COHERENT EXCITATION BY MAGNETIC RESONANCE

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INTRODUCTION

The transient behavior of spin-dependent transition rates of dipolar D and exchange J coupled paramagnetic (s=1/2) spin pairs, during coherent magnetic resonance excitation, is studied numerically using Liouville space formalism. We predict that the Rabi oscillation under the condition of strong D coupling will have a $\sqrt{2}\gamma B_1$ component, where γ is the gyromagnetic ratio and B_1 is the field strength of the driving radiation. In addition, we find that existing experimental pulsed electrically and optically detected magnetic resonance (pEDMR/pODMR) data can be modeled by the inclusion of a strong J, such that the difference in resonant Larmor frequencies within the pair (the so-called Larmor separation) $\Delta\omega$ is small compared to the difference of J and D. These results show that a pEDMR or pODMR experiment can measure exchange and dipolar coupling strengths between spin pairs.

THEORY

The evolution of the spin-pair density matrix ρ under coherent magnetic resonance is governed by the stochastic Liouville equation,[1]

$$\partial_t \hat{\rho} = \frac{i}{\hbar}[\hat{\rho}, \hat{H}\,] + S[\hat{\rho}].$$

H is the spin-pair Hamiltonian with Zeeman, exchange, and dipolar terms, and S is a stochastic term with annihilation and creation terms present during the excitation.

COMPUTATIONAL METHOD

We adopt a Liouville space formalism,[2] which leads to a factor of 10^3 decrease in computation time over conventional ODE solvers. The essence of the Liouville space formalism is representing the 4x4 spin-pair density matrix as a 16x1, and generating a set of 16x16 superoperators. Adopting this convention gives an extremely convenient solution to the inhomogeneous stochastic Liouville equation,

$$\hat{\rho}(t) = e^{Gt}(\hat{\rho}(0) + G^{-1}K) - G^{-1}K,\ \ \hat{\rho}(0) = G_s^{-1}K.$$

G is the evolution superoperator, G_s is the steady-state (B_1=0) evolution superoperator, and K is the generation term. From this, we generate a transient of the observable $Q(\tau)$. The observable does not depend on polarization (as in an ESR experiment), but rather depends on the permutation symmetry and transition rates of the spin pair. $Q(\tau)$ is then fast fourier transformed, making the Rabi frequencies of $Q(\tau)$ explicit.

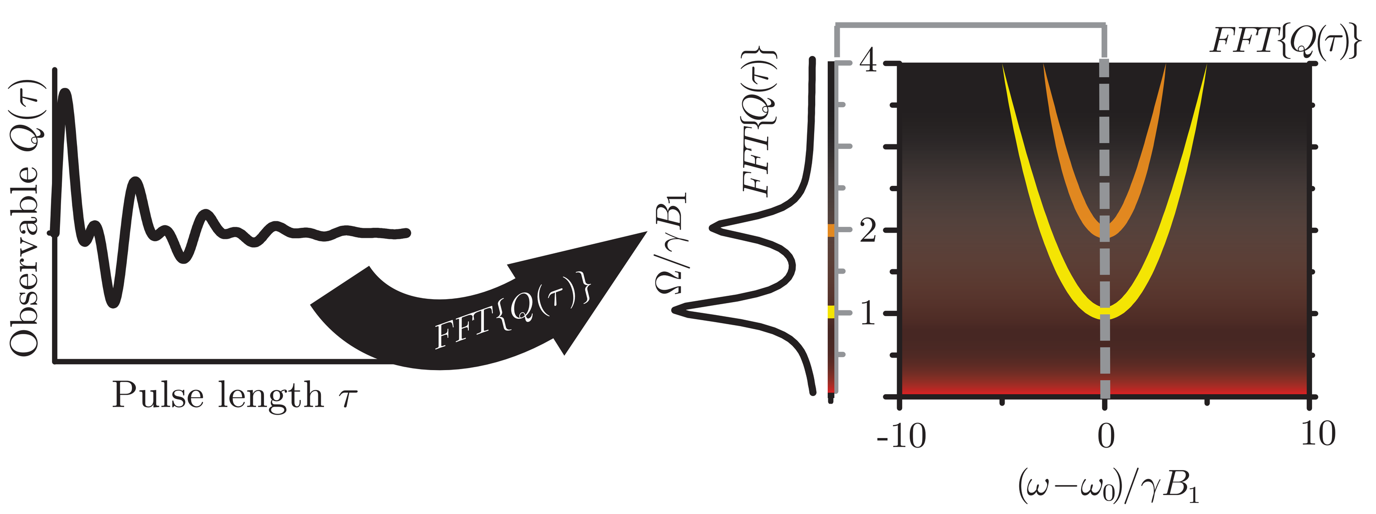

Fig. 1 A visual description of the computational method is given. First, parameters are selected and a transient of the observable $Q(\tau)$ is generated. Then, the transient is (fast) Fourier transformed. The frequency of the magnetic resonance pulse is swept and many transients are obtained, generating a Rabi mapping that is compared with experimental data.

RESULTS

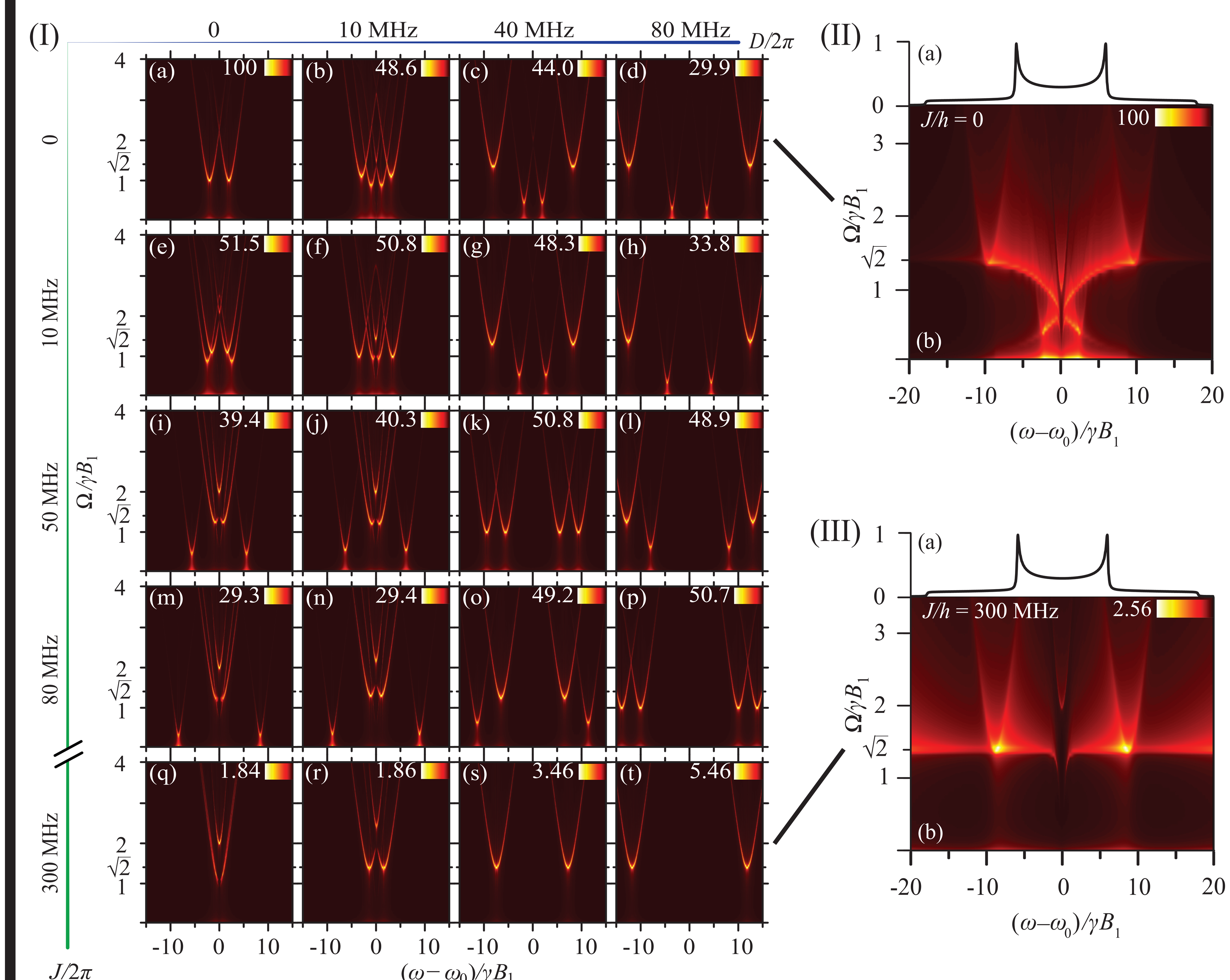

Fig. 2 Plots of the FFT of the observable $Q(\tau)$ as a function of driving frequency ω. ω_0 is the average of the Larmor separation between the spin pair, and Ω is the Rabi frequency. The color scales are in arbitrary units, with magnitudes listed next to the scale. $\Delta\omega$ = 40 MHz in all graphs. (I) A mapping comparing a wide range of dipolar D and exchange J couplings. (II) A dipolar Pake distribution is generated with J = 0 and D = 80 MHz. (III) A dipolar Pake distribution is generated with J = 300 MHz and D = 80 MHz.

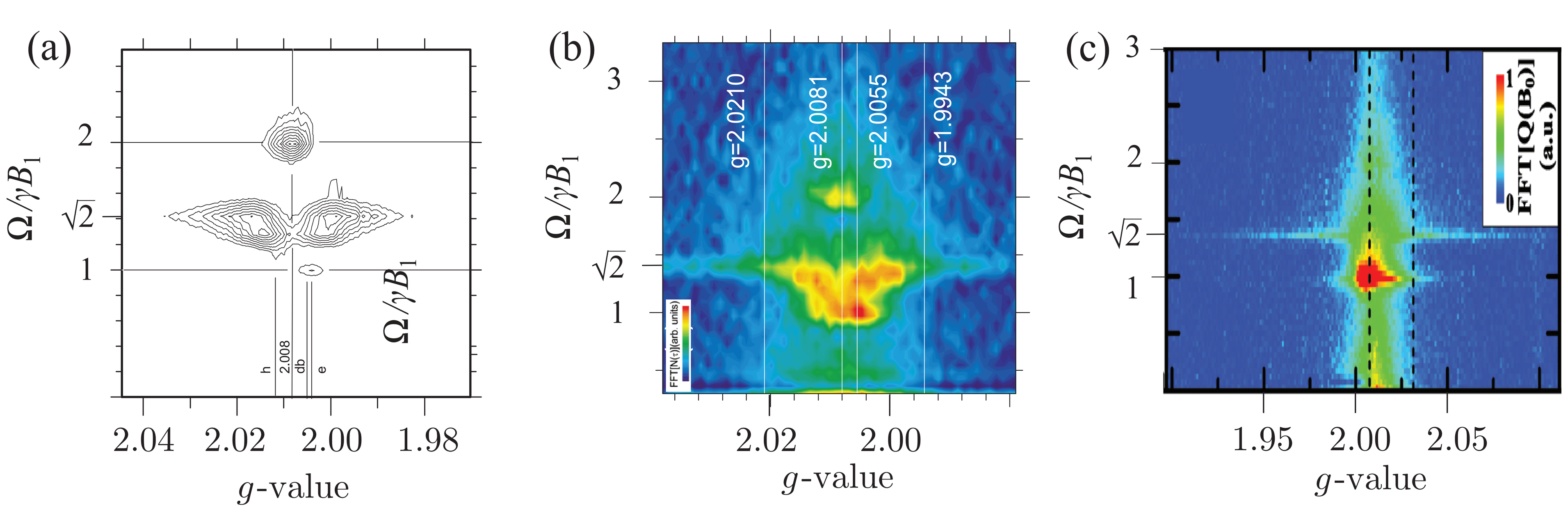

Fig. 3 (a-c) Adapted from Ref. [3-5], respectively. (a) Experimental pODMR data from amorphous silicon (a-Si:H). (b) Experimental pODMR data from geminate charge carrier pairs in a-Si:H. (c) Experimental pEDMR data from amorphous silicon-rich silicon-nitride.

DISCUSSION

The experimental data in Fig. 3a and 3b show a strong signal with Rabi frequency of $\sqrt{2}\gamma B_1$ and no strong low-frequency components ($\leq\gamma B_1$). However, Fig. 2 IIb, which has only dipolar coupling, shows strong low frequencies. In Fig. 2 IIIb, our simulation predicts that a spin pair with a large dipolar coupling and small Larmor separation $\Delta\omega$ compared to the difference of J and D will also produce a Rabi frequency of $\sqrt{2}\gamma B_1$ and no strong low frequencies. From this, we conclude that strong dipolar and strong exchange coupling account for the a-Si:H data.

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