

## Collective Coupling of a Macroscopic Number of Single-Molecule Magnets with a Microwave Cavity Mode

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(Received 22 July 2013; revised manuscript received 8 September 2013; published 26 March 2014)

We report the observation of strong coupling of a macroscopic ensemble of  $\sim 10^{16}$   $\text{Fe}_8$  molecular nanomagnets to the resonant mode of a microwave cavity. We use millimeter-wave spectroscopy to measure the splitting of the system's resonant frequency induced by the coupling between the spins and the cavity mode. The magnitude of this splitting is found to scale with  $\sqrt{N}$ , where  $N$  is the number of collectively coupled spins. We control  $N$  by changing the system's temperature and, thereby, the populations of the relevant spin energy levels. Strong coupling is observed for two distinct transitions between spin energy states. Our results indicate that at low temperatures nearly all of the spins in the sample couple with the cavity's resonant mode even though there is substantial inhomogeneous broadening of the  $\text{Fe}_8$  spin resonances.

DOI: 10.1103/PhysRevLett.112.120501

PACS numbers: 03.67.Lx, 42.50.Pq, 75.50.Xx, 76.30.-v

For two decades, single-molecule magnets (SMMs) have held promise to become an ultrahigh-density magnetic storage medium [1], and there has been significant recent progress towards realizing this goal [2]. In tandem, because SMMs are quantum systems [3], they have been suggested as possible qubits [4]. Quantum coherent phenomena have been observed in several SMM systems [5–8] with some exhibiting coherence times on the order of 1–10  $\mu\text{s}$ . There has also been major progress in addressing individual SMMs [9–16]. Alternatively, SMMs may be employed as a form of quantum memory within a hybrid quantum processor [17–21] in which quantum information is stored holographically in the entire crystal rather than bitwise in individual molecules. A first step towards implementing such a scheme with SMMs is to demonstrate that SMMs can quantum coherently couple to the photon states of a resonant cavity. Here we present spectroscopic evidence that nearly all of the  $\sim 10^{16}$  molecules in a crystal of the  $\text{Fe}_8$  SMM collectively couple with a resonant cavity mode. We find that the strength of the coupling scales with  $\sqrt{N}$ , where  $N$  is the number of collectively coupled spins, a parameter that depends on the system's temperature. Our results indicate that such collective coupling occurs even though there is substantial inhomogeneous broadening of the  $\text{Fe}_8$  spin resonances.

Coherent coupling between two-level systems (e.g., spins) and cavity photons lies at the heart of cavity quantum electrodynamics. Such interactions have been seen in many systems, including individual atoms [22], Bose-Einstein condensates [23], semiconductor quantum dots [24,25], and superconducting qubits [26]. In each of those systems the coupling is via electric-dipole transitions. Recently, coupling cavity photons to spins via magnetic dipole transitions has been investigated [27]. This weaker coupling, while more challenging to observe, may lead to longer coherence times. Coupling of spins and cavity photons

has been observed in several low-spin systems, including standard electron-spin resonance materials [28,29], nitrogen-vacancy centers in diamond [21,30,31], N-doped buckyballs [32],  $\text{Cr}^{3+}$  impurities in ruby [33], as well as other magnetically doped materials [32,34,35]. In all of these systems the spin belongs to a single atom, ion, or nucleus. In contrast, SMMs are more “macroscopic” artificial magnets where the spin degree of freedom is a joint property of an entire metal-oxide molecular cluster. These magnets have a large magnetic anisotropy, which produces a sizeable zero-field splitting between spin states. This enables coupling the spins to microwave photons at low magnetic fields, making some SMMs (with appropriate anisotropy values) potentially integrable with superconducting resonators and qubit circuits, which cannot operate at high fields. The macroscopic nature of SMMs presents a complication: For many SMMs, variations in the local environment of the molecules within a crystal lead to slightly different properties for each molecule [36] and inhomogeneous broadening of spectral resonance lines [37]. At the same time, with  $\sim 1$  molecule per unit cell and a large ( $s \sim 10$ ) magnetic moment, SMMs also have an extremely high spin density, leading to a much stronger spin-photon interaction than what is seen in many other spin systems. Our results show that the high spin density in SMMs can be harnessed to create a coupling strong enough to overcome the intrinsic inhomogeneity of the system.

The  $\text{Fe}_8$  molecule [Fig. 1(c)] is a spin-10 object with a large magnetocrystalline anisotropy, resulting in a preferred orientation for the spin, the so-called “easy” axis. As the spin tilts away from this axis, its energy increases, creating an effective double-well potential [Fig. 1(a)] as a function of the angle between the spin and the easy axis. The  $\text{Fe}_8$  spin Hamiltonian is given by

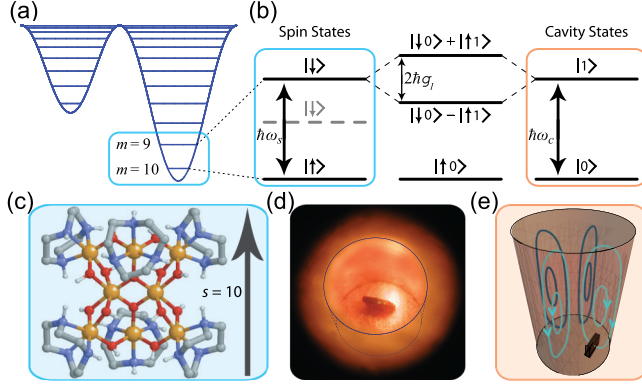


FIG. 1 (color online). (a) Double-well potential for a single-molecule magnet in a magnetic field. The levels correspond to different spin-orientation states. (b) Schematic of spin-photon interaction. One pair of SMM levels (the lowest two for the case shown) are labeled  $|\uparrow\rangle$  and  $|\downarrow\rangle$ , as shown. Photon-number states are labeled  $|0\rangle$  and  $|1\rangle$ . A magnetic field causes the excited state  $|\downarrow\rangle$  to shift upwards (from the gray, dashed level). When  $|\downarrow\rangle$  and  $|1\rangle$  are nearly degenerate, the two states hybridize, as shown in the middle of the panel, producing a minimum splitting of  $2\hbar g_1$ . (c) Structure of the  $\text{Fe}_8$  SMM studied. (d) Photograph of an  $\text{Fe}_8$  single crystal mounted in a cylindrical resonant cavity of radius 1.28 mm and depth 3.96 mm. Lines have been added to guide the eye. (e) Magnetic field lines for the  $\text{TE}_{011}$  resonant mode in the cavity.

$$H = DS_z^2 + E(S_x^2 - S_y^2) + C(S_+^4 + S_-^4) - g\mu_B \vec{B}_{\text{ext}} \cdot \vec{S}, \quad (1)$$

where  $\vec{B}_{\text{ext}}$  is the externally applied magnetic field,  $g \approx 2$ ,  $D = -25.2$  eV,  $E = -4.02$  eV, and  $C = 7.4 \times 10^{-4}$  eV [38]. The first term in Eq. (1) makes the  $z$  axis the easy axis and defines the double-well potential. The spin has  $2s + 1 = 21$  possible orientation states,  $m = -10, -9, \dots, 10$  [levels in Fig. 1(a)]. The easy-axis field component  $B_z$  tilts the potential, as shown, and increases the energy differences between the lowest states.

Because of its anisotropy, an SMM's energy levels are typically anharmonically dispersed, as shown in Fig. 1(a), and so only one pair of spin levels will effectively couple to the cavity at a time. This pair then behaves as an effective spin-1/2 system. We relabel the lower (upper) of the relevant states as  $|\uparrow\rangle$  ( $|\downarrow\rangle$ ) and define  $\hbar\omega_S$  as the energy difference between these two states. For example, when we truncate the states in Fig. 1(a) to the two lowest levels, we set  $|\uparrow\rangle = |m = 10\rangle$  and  $|\downarrow\rangle = |m = 9\rangle$ .

The interaction of a two-level system with a cavity can be modeled by the Jaynes-Cummings Hamiltonian [39]:

$$H = H_{\text{spin}} + H_{\text{rad}} + H_{\text{int}}, \quad (2)$$

where  $H_{\text{spin}} = (\hbar\omega_S/2)(1 - \sigma_z)$  is the spin Hamiltonian [i.e., Eq. (1) truncated to the two relevant levels],  $H_{\text{rad}} = \hbar\omega_C a^\dagger a$  is the Hamiltonian for the cavity mode, and  $H_{\text{int}} = (\hbar g_1/2)(a\sigma_- + a^\dagger\sigma_+)$  is the spin-photon

interaction Hamiltonian. The  $\sigma$ 's are standard Pauli spin matrices in the  $\{|\uparrow\rangle, |\downarrow\rangle\}$  basis and  $a^\dagger$  ( $a$ ) is the photon creation (annihilation) operator for the cavity mode, acting on photon-number basis states  $|n\rangle$ . Offsets have been chosen to make the energy of the  $|\uparrow\rangle|0\rangle$  ground state zero. The spin-radiation interaction strength  $g_1$  is given by

$$g_1 = |\langle \uparrow | S_T | \downarrow \rangle| g\mu_B B_{\text{rf}} / \hbar, \quad (3)$$

where  $B_{\text{rf}}$  is the radiative magnetic field of a single photon and  $S_T$  is the projection of the spin operator in the direction of  $B_{\text{rf}}$ . The subscript 1 in  $g_1$  indicates that a single spin is coupled to the cavity. With  $n \leq 1$ , the excited eigenstates of Eq. (2) are

$$\begin{aligned} |+\rangle &= \sin(\varphi/2)|\downarrow\rangle|0\rangle + \cos(\varphi/2)|\uparrow\rangle|1\rangle, \\ |-\rangle &= \cos(\varphi/2)|\downarrow\rangle|0\rangle - \sin(\varphi/2)|\uparrow\rangle|1\rangle, \end{aligned} \quad (4)$$

with energies

$$E_{\pm} = \frac{\hbar}{2}((\omega_C + \omega_S) \pm \sqrt{\Delta^2 + 4g_1^2}), \quad (5)$$

where  $\Delta = \omega_C - \omega_S$  is the cavity-spin detuning and  $\tan \varphi = 2g_1/\Delta$ . Equation (5) describes two branches of a hyperbola with asymptotes  $E = \hbar\omega_C$  and  $E = \hbar\omega_S$ . For large  $\Delta$ , the excited eigenstates approach the independent excitation of the cavity or the spin [dashed gray level  $|\downarrow\rangle$  in Fig. 1(b)], respectively, and the excited states of the total system are well described by  $|\uparrow\rangle|1\rangle$  and  $|\downarrow\rangle|0\rangle$ . Applying an external magnetic field increases  $\omega_S$ , raising the energy of the  $|\downarrow\rangle$  state [from dashed to solid level in Fig. 1(b)]. When  $\omega_S$  becomes close to  $\omega_C$  [vertical dashed black line in Figs. 2(b),(c)], the spin will interact with the cavity mode and the system's excited states hybridize. At  $\Delta = 0$  and with one excitation in the system, the splitting between the two branches,  $E_+ - E_-$  is  $2\hbar g_1$ , the vacuum-Rabi splitting, and the excited states of the system become simply  $|\pm\rangle = (1/\sqrt{2})(|\downarrow\rangle|0\rangle \pm |\uparrow\rangle|1\rangle)$ , the two split states in Fig. 1(b).

Using the structure of our cavity mode, it is straightforward to calculate the single-photon field at the position of the sample to be  $B_{\text{rf}} = 3.7(6) \times 10^{-7}$  G. Equation (3) then yields  $g_1/2\pi = 2.4$  Hz, much too small to be detected in a realistic experiment. As first shown by Tavis and Cummings [41], when  $N$  identical spins couple to the cavity, the interaction strength becomes

$$g_N = \sqrt{N}g_1. \quad (6)$$

The system's energies are still described by Eq. (5) with  $g_1$  replaced by  $g_N$  and the eigenstates by Eq. (4) with  $|\uparrow\rangle$  and  $|\downarrow\rangle$  replaced, respectively, by  $|\uparrow\rangle = |\uparrow\uparrow\uparrow\dots\uparrow\rangle$  and  $|\downarrow\rangle = (1/\sqrt{N})(|\downarrow\uparrow\uparrow\dots\uparrow\rangle + |\uparrow\downarrow\uparrow\dots\uparrow\rangle + \dots + |\uparrow\uparrow\uparrow\dots\downarrow\rangle)$ , where the latter describes an equal superposition of each spin being flipped into the excited state (while the remainder stay in the ground state). For the case relevant to our experiments, where

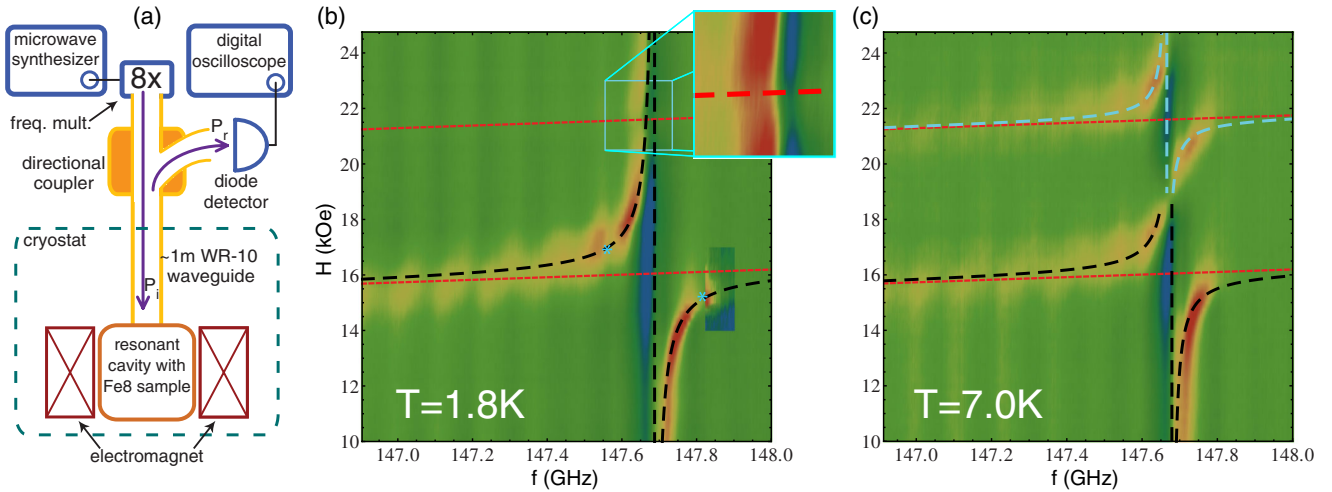


FIG. 2 (color online). Spectroscopic technique and results. (a) Schematic diagram of experimental apparatus. (b) and (c) Absorbed power as a function of magnetic field and radiation frequency at 1.8 and 7.0 K, respectively. Yellow and red indicate regions of significant power absorption. The dark blue regions are largely artificial, produced by our background-subtraction procedure [40]. The lower (upper) red dashed line is the Zeeman energy separation for the  $m = 10$ -to-9 (9-to-8) transition. The dashed black (cyan) curve is a fit of the data for the lower- (upper-)field data to Eq. (5). The outset of (b) shows a zoomed-in view of the boxed region with a slightly different coloring scheme. Similarly, the data within the lower boxed region in (b) are rendered with a different coloring scheme to emphasize the weaker features. In both (b) and (c), an essentially field-independent background was subtracted to enhance visual presentation [40].

$N \sim 10^{16}$  spins couple to a cavity mode,  $g_N/2\pi$  is  $\sim 200$  MHz, an easily detectable frequency splitting.

In coupling to the cavity, the collection of  $N$  spins behaves as one “superspin” with  $s = N/2$  [42]. The spin state  $|\downarrow\rangle$  corresponds to a rotation of the superspin vector from the  $z$  axis (such that the  $z$  component of the spin is reduced by 1). In our experiment, the number of photons in the cavity  $n$  is on the order of  $10^{10}$ . Nevertheless, Eq. (6) remains valid when the assumption  $n \leq 1$  is replaced by the less stringent condition  $n \ll N$ . The latter corresponds to the limit in which the superspin’s angle relative to the  $z$  axis remains small. The anharmonic limit, in which this angle is large, gives rise to superradiant states [42]. In practice,  $N$  depends on temperature and thereby permits *in situ* control of the coupling strength  $g_N$ .

Crystals of  $\text{Fe}_8$  were synthesized using standard techniques. The crystal used for measurements was photographed under a microscope to determine its dimensions. Using those and the known unit cell for  $\text{Fe}_8$  [43], we determined that the sample consists of  $N_0 = 2.3(4) \times 10^{16}$  SMMs. Figure 1(d) shows a photograph of a single crystal of the  $\text{Fe}_8$  SMM mounted in our cylindrical copper cavity. The  $\text{TE}_{011}$  mode of our cavity has a resonant frequency of 147.677(2) GHz and  $Q \sim 4000$ . For this mode, the oscillating magnetic field, shown in Fig. 1(e), is nearly perpendicular to the easy axis. The sample is mounted with its easy axis  $\theta \sim 35^\circ$  from the external dc magnetic field, which is parallel to the cavity’s symmetry axis. Our experimental setup is shown schematically in Fig. 2(a). We performed measurements of the radiation power reflected from the cavity-sample system as a function of frequency and dc magnetic field at several temperatures between  $\sim 1.8$  and 20 K.

Figures 2(b) and 2(c) show absorbed power at 1.8 and 7.0 K, respectively, for a range of frequencies and magnetic fields. Resonances of the system appear as yellow or red regions. The data exhibit two distinct resonant branches, each of which corresponds to one of the coupled spin-photon states in Eq. (4). At low magnetic fields, the resonances appear near the bare cavity resonance frequency (vertical dashed line) and the excitation frequency for the dipole-allowed  $m = 10$ -to-9 spin transition (lower red dashed line). When the field approaches the value at which these resonances would cross in the absence of interaction, a clear avoided crossing opens up with the upper-left branch curving and eventually approaching the cavity resonance frequency. The lower right branch tends towards the spin transition frequency but signal strength is lost as the frequency increases. Irrespective of this loss of signal, the upper left branch clearly transits smoothly between cavitylike and Zeeman-like behavior, indicating that the system has entered the so-called strong-coupling regime with states like those described by Eq. (4). Moreover, we find that there is a range of fields at which two resonance frequencies are observed [40], a telltale sign of strong coupling. Both branches can be fit very well by Eq. (5) (with  $g_1$  replaced by  $g_N$ ) as shown by the black dashed curves in Fig. 2(b) [40]. Only two parameters in the fit are unconstrained by the spin Hamiltonian or the cavity’s resonant frequency:  $\theta$ , the angle between the easy axis and the magnetic field, a parameter that was restricted to be the same at all temperatures, and  $g_N$ , which was allowed to vary with temperature. The former determines the slope of the spin transition frequency’s field dependence (lower red dashed line) while the latter determines the gap between the

two branches of the hyperbola. Our fits provide a best value of  $\theta = 37.7^\circ$ , close to the expected value of  $35^\circ$  based on the sample's orientation. For the data shown in Fig. 2(b), we obtain  $g_N/2\pi = 0.519(4)$  GHz.

Figure 2(b) shows another, much smaller feature where the upper red dashed line, corresponding to the  $m = 9$ -to-8 transition, intersects the cavity frequency; the feature is highlighted in the inset. Since at 1.8 K there are far fewer molecules in the excited  $m = 9$  state than in the  $m = 10$  ground state, the value of  $N$  for the 9-to-8 transition is very small, resulting in a smaller coupling to the cavity mode. The splitting  $g_N$  for this transition can be increased by raising the temperature  $T$  and, thereby,  $N$  for the  $m = 9$  state. Indeed, as shown in Fig. 2(c), increasing  $T$  to 7.0 K decreases the magnitude of the splitting associated with the (lower field) 10-to-9 transition and dramatically increases the coupling associated with the (higher field) 9-to-8 transition. This observation reflects the fact that raising the temperature monotonically reduces the population in the ground ( $m = 10$ ) state while initially increasing the population of the excited ( $m = 9$ ) state.

We fit the data in Fig. 2 (and similar data at other temperatures) to obtain values of  $g_N$  for each spin-cavity resonance at all temperatures for which a good fit to Eq. (5) could be obtained.  $g_N^2$  should be proportional to the relative population  $p = N/N_0$  for the relevant transition. In Fig. 3, we plot  $(\hbar g_N / |\langle \uparrow | \vec{S}_T | \downarrow \rangle| \mu_B g)^2 = p N_0 B_{rf}^2$  as a function of temperature for the two transitions measured. It is straightforward to calculate the populations  $p$  of the  $m = 10$  and 9 states as a function of temperature with no adjustable parameters [40]. The solid curves in Fig. 3 show this temperature dependence for the relevant levels,  $m = 10$  and  $m = 9$ . The agreement between the data and the corresponding populations is evident. The only adjustable parameter for these curves is the product of  $N_0$  and  $B_{rf}^2$ , which determines the vertical scale of the curves. Taking  $N_0 = 2.3 \times 10^{16}$ , we determine  $B_{rf} = 5.30(1) \times 10^{-7}$  G for the 10-to-9 transition [Fig. 3(a)] and  $4.98(3) \times 10^{-7}$  G for the 9-to-8 transition [Fig. 3(b)]. These values agree well with each other and are on the same order as our calculated value of  $3.7(6) \times 10^{-7}$  G. The discrepancy may arise from modal mixing with the nearly degenerate  $TM_{111}$  mode.

Inhomogeneous broadening in  $Fe_8$ , as in many SMMs, arises from variations within a sample of the anisotropy parameter  $D$ , as well as other Hamiltonian parameters [37]. The broadening can be seen in the rather wide spin resonances in the data in Fig. 2, which have a Gaussian width of  $\sim 760$  Oe [40], corresponding to a frequency width of  $\sigma_\omega/2\pi \sim 1.7$  GHz. A Gaussian distribution of  $N$  spin resonant frequencies  $\omega_S$  is expected to still couple collectively if  $\sigma_\omega \lesssim g_N$  [44–46]. Remarkably, our results do not quite meet this condition with  $\sigma_\omega \gtrsim 3g_N$ . In this regime, theory predicts a single S-shaped curve centered on the intersection of the Zeeman line with the cavity frequency (see [40]) instead of the

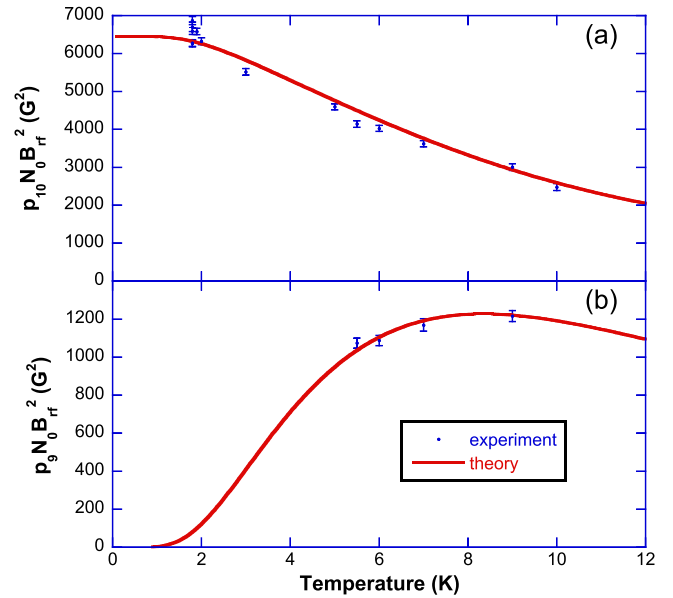


FIG. 3 (color online). Measured frequency splitting as a function of temperature for the (a)  $m = 10$ -to-9 transition and (b)  $m = 9$ -to-8 transition. The splitting has been recast in terms of the relative level population  $p$  of the lower level (see text) and compared with populations calculated using the known energy levels for the  $Fe_8$  single-molecule magnet (solid curves).

two distinct branches that we observe in Figs. 2(b),(c). The light blue asterisks in Fig. 2(b) indicate the turning points of the predicted resonance curve, calculated following [45] and using the measured value of  $\sigma_\omega$  and the value of  $g_N$  determined for that data set. The fact that the actual data do not show such turning points (most obvious in the left branch) indicates an unexpected collective coupling despite significant inhomogeneous broadening. This may be due to the existence of a small nonlinearity in the spin-cavity or spin-spin interactions that induces the spins to synchronize [47] as  $\Delta$  becomes small, or the presence of some weak additional coupling mechanism, perhaps mediated by the crystal lattice [48]. These and other possible mechanisms are the subject of ongoing experimental and theoretical investigations. Regardless of the specific mechanism, our findings indicate that the spins need not have nearly identical resonant frequencies in order to couple collectively to a cavity mode but can do so even with substantial inhomogeneous broadening.

We thank M. Bal for much useful advice and for his work on an earlier version of this experiment. We also thank E. M. Chudnovsky, J. A. Grover, D. S. Hall, D. B. Haviland, S. Hill, A. J. Millis, L. A. Orozco, M. P. Sarachik, D. I. Schuster, S. H. Strogatz, F. K. Wilhelm, and D. J. Wineland for useful discussions, and R. Cann for technical assistance and advice. Support for this work was provided by the National Science Foundation under Grants No. DMR-0449516, No. DMR-1006519, and No. DMR-1310135 and by the Amherst College Dean of Faculty.

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