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Off-resonant manipulation of spins in diamond via precessing magnetization of a proximal ferromagnet

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We report the manipulation of nitrogen vacancy (NV) spins in diamond when a nearby ferrimagnetic insulator, yttrium iron garnet, is driven into precession. The change in NV spin polarization, as measured by changes in photoluminescence, is comparable in magnitude to that from conventional optically detected magnetic resonance, but relies on a distinct mechanism as it occurs at a microwave frequency far away from the magnetic resonance frequency of the NV spin. This observation presents an approach to transferring ferromagnetic spin information into a paramagnet and then transducing the response into a robust optical signal. It also opens strategies for studying ferromagnetism and spin transport at the nanoscale.

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Understanding the transport of spin and energy between dissimilar materials is a topic of intense current interest, reflecting both the scientific richness of the topic as well as its technological potential [1–7]. Metal/metal interfaces have been extensively studied and, to a lesser degree, so have metal/semiconductor and metal/insulator systems. However, the transfer of angular momentum between two insulating materials has been more challenging to study due to the lack of suitable detection methods.

Nitrogen vacancy (NV) centers in wide-band-gap insulating diamond provide an exceptional platform for performing spin-based measurements. The paramagnetic NV center is optically active, and its photoluminescence (PL) is dependent on the relative occupation of the lowest-lying electronic spin state of the defect center [8,9]. This enables optical measurement of the NV-center spin state with excellent sensitivity, making optically detected magnetic resonance of NV centers an area of intense research activity [10–22]. However, work done so far relies on manipulation of NV spins using magnetic resonance.

Here we present experimental evidence that the NV-center state can be modified nonresonantly (i.e., by irradiation with microwave magnetic fields at frequencies far from NV-center Larmor frequencies) by coupling to the dynamics of a proximal ferromagnetic insulator. This change can be detected as a change in the NV-center PL, as is done for conventional microwave-driven resonant spin manipulations. This effect promises valuable insights into interactions between spins in adjacent dissimilar materials. This is a particular case in which both materials are insulating and the interaction is effective over long distances (>300 nm). More generally, this provides a method for manipulating NV-center spins and could enable sensitive spatially resolved imaging of ferromagnetic phenomena by means of atomic NV centers [10,13,16].

Yttrium iron garnet (YIG), $Y_3Fe_5O_{12}$, was chosen for this experiment as a well-known ferrimagnetic insulator with exceptionally low damping [23,24]. Here an epitaxial YIG film, 20 nm thick, was grown on a gadolinium gallium garnet (GGG) (111) substrate by off-axis sputtering [25–27]. Continuous-wave microwave fields are applied to the sample by means of a 300-nm-thick and 30-μm-wide silver microstrip line patterned on top of the YIG. Nanodiamonds, 50–200 nm in size [as shown by scanning electron microscopy (SEM) image analysis] and containing up to a few thousand NV centers each, were dispersed on top of the lithographically defined microstrip line, as shown in Fig. 1. Atomic force microscope measurements indicate that the nanodiamonds form a 500-nm-thick film. Photoluminescence is excited in the NV centers using a 532 nm laser beam and is collected by a photodiode. A lock-in measurement is performed on the photodiode signal by modulating the amplitude of the applied microwave field.

The lock-in measurement of the resulting modulation of the PL intensity is presented in Fig. 2 as a function of an applied in-plane magnetic field and the applied microwave frequency. Data for a control sample with nanodiamonds on a GGG substrate without YIG is shown in Fig. 2(a). We observe the intrinsic and well-known magnetic resonances of the NV-center ground and excited states, starting at 2.87 and 1.43 GHz, respectively. Shown in Fig. 2(b) are the same data overlaid with the theoretically expected resonance conditions for the NV centers, which are obtained by solving the NV Hamiltonian in the presence of magnetic field parallel and perpendicular to the NV axis (see supplementary information of [10]). The features seen in the PL below 1.25 GHz [Fig. 2(a)] occur when the harmonics from the microwave synthesizer match the ground-state resonances (see Supplemental Material [28] for more information).

The data obtained from the nanodiamonds on top of the YIG film are shown in Fig. 2(c). The key difference between Figs. 2(a) and 2(c) is the feature in Fig. 2(c) that extends up from the lower left-hand corner. This is highlighted in Fig. 2(d) where the data from Fig. 2(c) is overlaid with a solid blue curve, showing the YIG ferromagnetic resonance condition. The blue dots show the YIG resonance condition as measured by reflected microwave power. These data are fit (blue curve) using the equation for the uniform ferromagnetic resonance (FMR) mode in a thin film [29], and we obtain a magnetization $\mu_0 M_s$ of 183 mT (see Supplemental Material [28] for more information). As can be seen, the intensity of the PL from the

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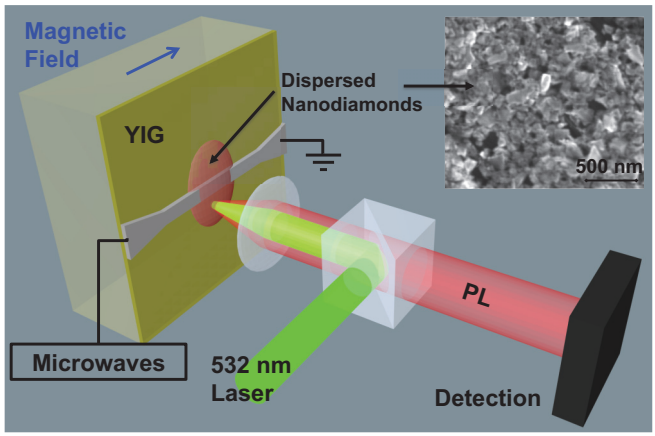

FIG. 1. (Color online) Experimental schematic: The sample is a 20-nm-thick single-crystal YIG film with nanodiamonds dispersed on top with a thickness of about 500 nm. To apply microwave fields to the sample, a silver microwire is patterned on the YIG. Green laser light is focused onto nanodiamonds near the wire, and the intensity of the resulting photoluminescence from the NV centers is measured. The inset is an SEM image of dispersed nanodiamonds.

NV centers strongly changes precisely when the YIG FMR is excited.

To test the robustness of this phenomenon, we have measured the microwave and laser power dependence of the FMR-related feature. The magnitude of this feature, as measured by the area under the peak, is presented in Fig. 3(b). This area was calculated for frequency sweeps [Fig. 3(a)] performed at a field of about 2.5 mT, where the FMR feature is well separated from any NV resonances. The data show that the feature can be measured in a broad window of experimental parameters and that the magnitude is almost linear in both laser and microwave power, within our measurement range. Also, while we have studied one spot in one sample in greater detail and are presenting those results here, we have seen this feature in all of the YIG samples that we have measured. These include samples grown by both sputtering and liquid phase epitaxy, having different thicknesses. We have also consistently found it over multiple spots within various samples, where the nanodiamond density is varying. The amplitude of the effect varies between samples and spots within a sample, but is consistent within the variability in microwave fields expected in these measurements. We also note that the increased amplitude of the NV excited-state peak in Fig. 2(c) as compared to Fig. 2(a) most likely results from differing microwave field experienced in the two samples as a consequence of variability in the microwave antenna fabrication (e.g., lithography or wire contacts) and is not linked to the YIG FMR.

We have ruled out numerous possible explanations for the observed behavior. We have verified that the harmonics of the microwave source, which cause low-frequency features in Fig. 2(a), are not causing the FMR-related peak. More details are presented in the Supplemental Material [28], where we show a comparison of the signal with and without a microwave low-pass filter. We have considered the effect of heating, caused by the FMR absorption in YIG, on the PL

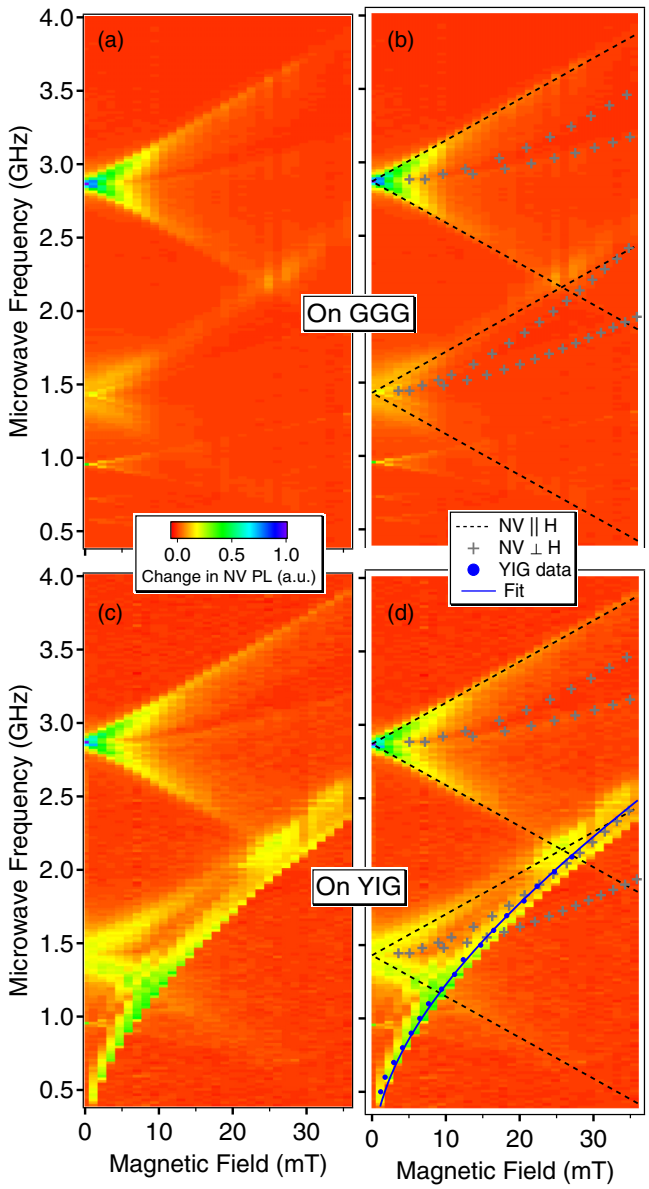

FIG. 2. (Color online) NV PL data while YIG undergoes FMR: (a) Raw data showing the change in the intensity of the PL from the NV centers in nanodiamonds dispersed on top of GGG, with no YIG, as a function of microwave frequency and magnetic field. (b) The same data overlaid with theoretical resonance conditions for the NV centers. The black dashed lines show the resonance condition for an NV center with the magnetic field parallel to the NV axis for the ground and excited states. The gray crosses show the resonance condition for an NV center with the magnetic field perpendicular to the NV axis. (c) Similar data from nanodiamonds dispersed on the YIG sample with the distinct feature corresponding to the YIG FMR condition. (d) The data from (c) with the FMR peaks measured using reflected microwave power (blue dots). Also shown is a fit (blue line) to the calculated dispersion relation for YIG-film FMR with the magnetic field in plane (see main text).

of NV centers. Several control experiments and estimates of the possible effect render this potential explanation for the FMR-induced feature highly unlikely. More details can be found in the Supplemental Material [28]. The linear response

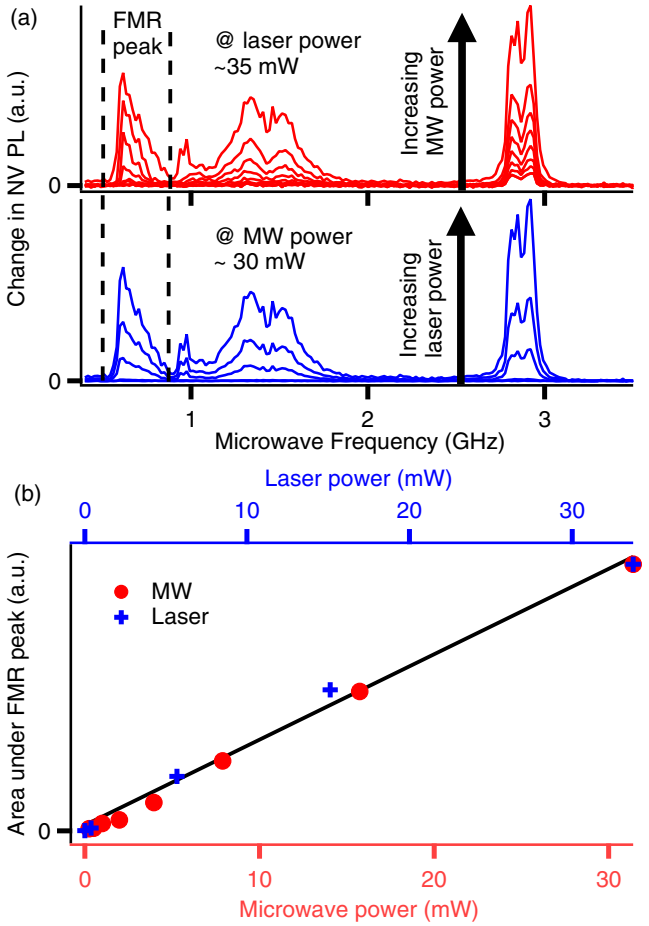

FIG. 3. (Color online) Power dependence of the FMR-induced feature: (a) Frequency sweeps at a magnetic field of 2.36 mT for various microwave (top panel) and laser (bottom panel) powers. (b) The area under the FMR peak [identified in (a), between the dashed lines] is shown as a function of the microwave (red dots, bottom axis) and laser (blue crosses, top axis) powers. A linear guide to the eye is provided which indicates that the FMR-induced feature has a fairly linear response to both laser and microwave power in our measurement range.

to microwave power rules out nonlinear effects such as second harmonic generation in the YIG, resulting in microwaves at the NV resonances, as a likely cause. It is unlikely that the YIG resonance emits microwaves strong enough to cause NV spin excitations at the far tails of the NV Lorentzian absorption spectrum. This would require microwave fields that are 20 times stronger than those applied with the strip line in order to obtain the observed signal magnitude at 0.5 GHz. Such enhancement is unlikely and should result in a much larger signal at the intersection of the NV excited state with the FMR feature. Furthermore, the NV PL away from resonance is virtually zero at all microwave powers, even in the GGG sample, indicating the difficulty in exciting these Lorentzian tails. The FMR-induced feature seems to be resulting from an as-yet unrecognized mechanism for transferring angular momentum from ferromagnets into NV centers.

There are two key points to note about the FMR-induced feature in the PL signal. First, it is seen at frequencies and fields well separated from the NV center's own resonance conditions. This is in clear contrast to the quantum computing and magnetometry techniques being developed, where the spin state of NV centers is coherently manipulated by microwave fields meeting the magnetic resonance conditions [10–21]. Instead, we see a change in the PL that correlates to the excitation of the YIG magnetization into precession by means of ferromagnetic resonance. It is remarkable that excitations at energies as much as three to six times smaller than any NV-center resonance have such a large effect on the NV-center spin state. Second, the FMR-induced feature is comparable in amplitude to the intrinsic NV resonances. The large amplitude of the signal implies that a significant number of NV centers in our laser spot must be contributing to the signal. This suggests that since the nanodiamond film is 500 nm thick, the coupling must be either long range (extending hundreds of nanometers) or that spin transport by means of spin diffusion plays a role [30].

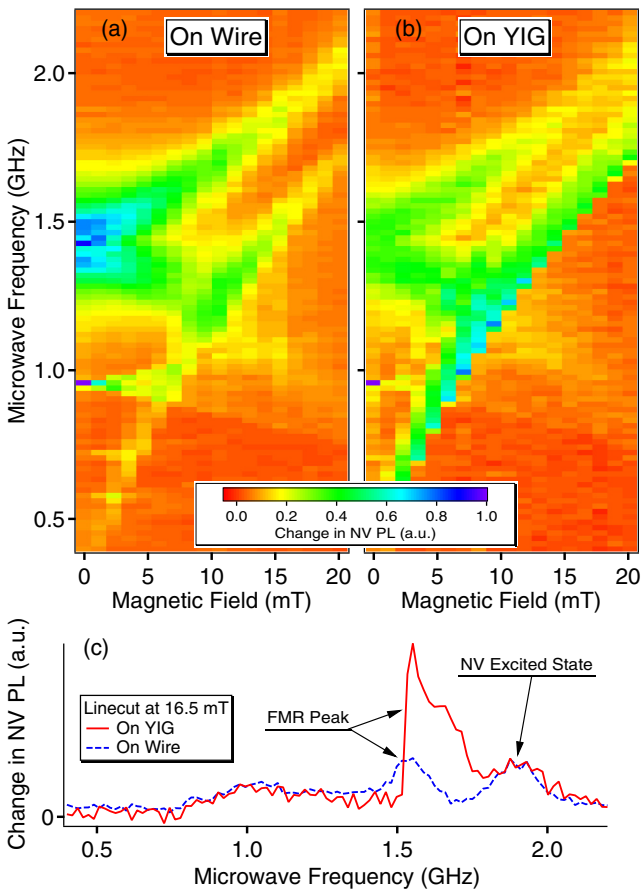

FIG. 4. (Color online) NV-center PL data on YIG and on wire above YIG: (a) Data taken with the laser spot focused on top of the ~300-nm-thick patterned microwire. The data shows that the signal corresponding to the YIG FMR is still present, though reduced. The coupling must extend at least 300 nm through the wire. (b) Data from the nanodiamonds directly on top of the YIG for comparison. (c) Line cuts of the data in (a) and (b) at 16.5 mT. The FMR-induced peak is reduced by three times relative to the NV excited-state peak for the case of nanodiamonds on top of the microstrip, compared to the case directly on YIG.

To probe the spatial extent of the coupling, we repeated the measurement with our laser spot focused on the nanodiamonds on top of the microstrip line, where the nanodiamonds are separated from the YIG by more than 300 nm. This data can be seen in Fig. 4(a) and compared to the signal when the diamond is directly on top of the YIG in Fig. 4(b). Line cuts (at 16.5 mT), presented in Fig. 4(c), show that the FMR-induced feature is reduced but clearly persists even when the nanodiamonds are not in direct contact with the YIG.

While a clear explanation for the effect is not forthcoming from our experimental results, we can make a few observations. First, the insulating nature of both materials rules out long-range carrier-mediated transport of spins. Second, one of the unique aspects of this experiment is that both the YIG magnetization and the NV-center spins are out of equilibrium when the YIG is on FMR: YIG due to its resonance and the NV centers due to the hyperpolarizing action of the laser excitation [31,32]. This is in contrast to other systems where angular momentum is transferred between spin subsystems, e.g., dynamic nuclear polarization or spin pumping [1,2,33], where spins in one spin subsystem relax by transferring polarization to another spin subsystem. The transfer of angular momentum from the YIG to the NV centers could result in relaxation towards equilibrium of both the spin systems, and thus be highly desirable for the overall system.

This phenomenon offers the opportunity of probing spin transport in the absence of conductors and employing an all-optical readout. These advantages dramatically reduce the potential confounding factors encountered in studies of spin transport in metallic systems. Thus this system offers an attractive and powerful approach to better understanding microscopic details of relaxation and spin transport in magnetic heterostructures. Experiments are underway on other ferromagnets and structures to gain more insight on this effect and how angular momentum is transferred between spin systems.

In summary, we have shown that the NV-center spin state can be manipulated by a coupling to the dynamic magnetization of YIG. The availability of a wide selection of ferromagnetic materials and structures (e.g., saturation magnetization and anisotropies) potentially offers a high degree of control in manipulating the NV spin state. The potential for ultra-high-resolution imaging of ferromagnetic phenomena using individual NV centers can have a significant impact as well. It should influence the fields of spintronics and quantum information by combining the sensitivity of the NV center and the tunability and scope of ferromagnetism.

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