Observation of a Novel Magnetic Transport Effect: Magnetization Transfer via a Combination of Spin Diffusion and Quantum Solid Diffusion

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Using NMR, we are studying a system in which spin diffusion, quantum solid particle diffusion, and quantum driven surface-substrate coupling contribute to magnetization transfer across a finite distance. In a mixture of polystyrene microspheres, Teflon microspheres, and a solid ^{3}He layer, we observe magnetization transport from ^{1}H through ^{3}He to ^{19}F nuclear Zeeman reservoirs. Experiments are conducted between 125 and 750 mK, in magnetic fields of 0.15 and 0.25 T, and over a wide range of ^{3}He coverages.

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We describe the idea behind the experiment by the following analogy. Consider the heat transfer between two spheres of differing materials in contact with each other. Both thermal impedance mismatching and small contact area will create a large thermal boundary resistance and thus poor heat transfer. However, if the materials were coated with a thin metal film, heat could be transferred between the two substrates via the film. The important parameters in this problem would then be the bulk heat capacities and diffusion constants as well as the thermal boundary resistance between each insulator and the metal film. We can study this system by putting a heat pulse in one substrate and monitoring the temperature changes in the film or the other substrate.

Our experiment is the magnetic analog of the above heat transfer experiment. Our substrates are polystyrene (^{1}H reservoir) and Teflon (^{19}F reservoir), and our film is a solid monolayer of ^{3}He. In the substrates, spin diffuses via the dipole-dipole interaction between fixed atoms ("spin diffusion"). At the interfaces between the ^{3}He and the substrates, there is surface magnetization transfer [1–9] (discussed below). In the ^{3}He layer, spin diffuses via quantum solid diffusion of the ^{3}He atoms ("spin convection"). We estimate this diffusion constant by Ja^2 where J is the exchange frequency and a is the distance between neighboring ^{3}He nuclei. Exchange frequencies between 5 [10] and 20 MHz [1] imply that the ^{3}He atoms diffuse 1 to 2 μm in 1 sec. Finally, we probe the system by flipping the spins of one substrate and monitoring the magnetization of the ^{3}He film or the other substrate.

Figure 1 shows the novel magnetization transport effect. The figure shows the ^{19}F magnetization response to a 120° proton pulse when a ^{3}He monolayer covers the microspheres. (In units of nuclear magnetons, the nuclear magnetic moments of ^{1}H, ^{19}F, and ^{3}He are 2.792, 2.627, and −2.127, respectively.) The proton pulse creates a magnetization gradient. For the first 45 sec, magnetization flows from the ^{19}F, through the ^{3}He, and to the ^{1}H. In particular, bulk ^{19}F magnetization spin diffuses to the surface of a Teflon microsphere. At the Teflon surface, magnetization is transferred from the ^{19}F nuclei to the ^{3}He nuclei via the ^{3}He exchange driven coupling [1–9] (discussed below). These ^{3}He nuclei then diffuse via quantum exchange to a polystyrene microsphere. At the polystyrene surface, magnetization is transferred from the ^{3}He nuclei to the ^{1}H nuclei. Finally, magnetization from the surface ^{1}H nuclei spin diffuses to the bulk ploystyrene. The nuclear species equilibrate among themselves faster than they equilibrate with the refrigerator via the ^{3}He lattice (discussed below). After the first 45 sec of internuclear thermalization, however, we see thermalization with the lattice taking place. Eventually the ^{19}F nuclei recover to their equilibrium value of 1.0. These results are the first observation of a system in which spin diffusion, quantum solid particle diffusion, and quantum driven surface-substrate coupling contribute to magnetization transfer across a finite distance.

We now give a fuller account of the ^{3}He to substrate coupling. First, consider relaxation in the ^{3}He. ^{3}He atoms in the solid monolayer undergo quantum exchange, and this motion provides the time dependent homonu-

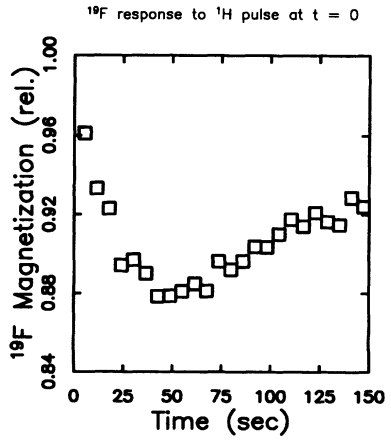

FIG. 1. The figure demonstrates the novel magnetization transport effect: the coupling of the spatially separated ^{19}F and ^{1}H reservoirs via a solid ^{3}He monolayer. The temperature is 485 mK and the field is 0.25 T.

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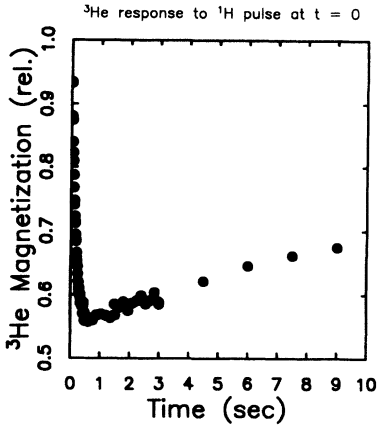

FIG. 2. The figure shows the cross polarization of ^{3}He nuclei by protons. The ^{3}He coverage is five monolayers. The temperature is 125 mK and the field is 0.15 T.

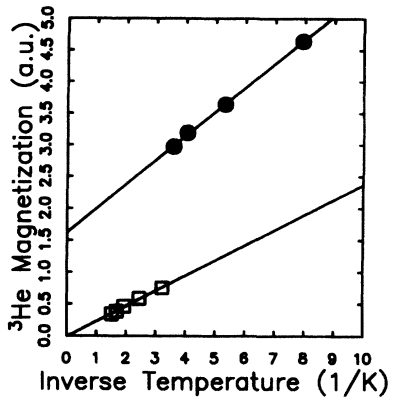

FIG. 3. The figure shows the ^{3}He equilibrium magnetization versus the inverse temperature at two coverages. From these data, we determine the amounts of liquid and solid ^{3}He in the cell. The lower line (□) corresponds to a solid ^{3}He monolayer. The upper line (•) corresponds to $1\frac{1}{2}$ Curie-like layers and $3\frac{1}{2}$ liquid layers.

clear dipole-dipole coupling responsible for surface ^{3}He relaxation. As more ^{3}He is added, liquid layers form. However, the surface relaxation continues to be the dominant relaxation path in the system. Surface magnetization is relaxed in the solid layer, and then magnetization is transferred from the liquid to the solid for further relaxation. Hence, the bulk ^{3}He relaxation is driven by surface ^{3}He motions [1,10–12].

Next, we consider the effect of surface ^{3}He motions on substrate relaxation. The mobile surface ^{3}He atoms couple to the hydrogen atoms on the polystyrene surface and thus provide a time dependent heteronuclear dipole-dipole coupling. One of the consequences of this coupling is that a ^{1}H nucleus and a ^{3}He nucleus can undergo a mutual spin flip with the Zeeman energy difference being picked up by the ^{3}He lattice. Hence the ^{1}H system can cross polarize the ^{3}He system. Alternatively, one can view these processes as the ^{3}He system relaxing the substrate system. Surface hydrogens are relaxed and then the magnetization in the bulk polystyrene spin diffuses to the surface for further relaxation by the ^{3}He. An analogous cross relaxation occurs between the liquid ^{3}He and the Teflon [1–9]. Furthermore, cross relaxation of a substrate by surface ^{3}He atoms has been demonstrated for a variety of diamagnetic insulating materials and has been suggested as a means for cooling them and characterizing their surfaces [6]. Figure 2 shows our results for the cross polarization of ^{3}He nuclei by protons. The figure shows the response of the ^{3}He magnetization to an arbitrary proton pulse (67°) at $t = 0$. The data are taken at 125 mK, 0.15 T (6.2 MHz), and a coverage of five monolayers. 1.0 is the ^{3}He equilibrium magnetization. The initial suppression time is of the order of the ^{3}He T_1 (0.3 sec), and the recovery time is of the order of the ^{1}H T_1 (27 sec). These results are consistent with previous studies involving ^{19}F based substrates. The authors give a quantitative discussion elsewhere [9].

The experimental setup is as follows. We pack the polystyrene Teflon mixture in the base of a test tube epoxied to a copper flange. We mount this sample cell, shown elsewhere [9], to the base of a dilution refrigerator. An electromagnet external to the cryostat produces the static NMR fields.

Compared to earlier work [9], we have made improvements in mixing the substrates. The 0.2 μm Teflon microspheres come in a powered form. The 0.2 μm polystyrene microspheres, however, come suspended in a surfactant. First, we ultracentrifuged the polystyrene colloid. Next, we drained off the liquid and mixed the residue with deionized water. We then centrifuged and drained off the liquid again. After the residue dried, we ground it with the Teflon, added methyl alcohol, and applied ultrasound for 30 min. We then put the slurry in an evacuated drying oven at 68 °C to boil off the alcohol. After grinding the resulting substance, we put it in the sample cell and flushed repeatedly with nitrogen and then ^{3}He at 50 °C.

We use NMR for thermometry and ^{3}He surface characterization. The NMR spectrometer is described in chapter 4 of Ref. [13]. We use a hydrogen-free ceramic paste to pot bare copper wire for the receiver coil. (This paste, produced by Aremco, is designed to hold at 2000 °C, but it works fine at 125 mK as well.) We check thermometry by calibrating the ^{19}F equilibrium magnetization versus $1/T$ where T is the temperature measured with a ^{3}He melting curve thermometer.

The amounts of liquid and solid ^{3}He in the cell can be computed from the ^{3}He equilibrium magnetization versus $1/T$. Figure 3 shows these data at two coverages. The lower line is Curie-like. At this low coverage, the sample loses thermal contact with the refrigerator at temperatures below 300 mK. The upper curve is also linear in $1/T$, but it has a nonzero offset. This is the temperature independent contribution of ^{3}He in the Fermi liquid regime. We compute the number of liquid and Curie-like

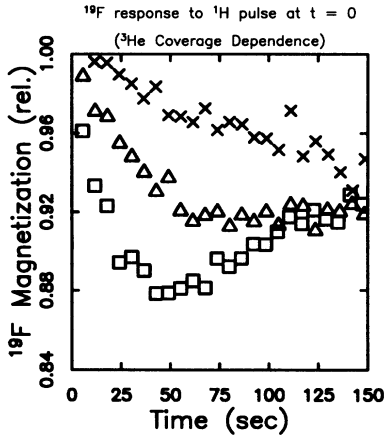

FIG. 4. The figure demonstrates the ^{3}He coverage dependence of the ^{19}F-^{1}H coupling. Data are taken at much less than a monolayer (×), a solid ^{3}He monolayer (□), and a cell full of liquid ^{3}He (△). The temperature is 485 mK and the field is 0.25 T.

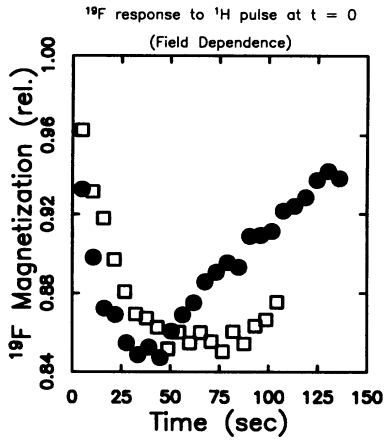

FIG. 5. The figure demonstrates the field dependence of the ^{19}F-^{1}H coupling at a ^{3}He coverage of five monolayers. Data are taken at 0.15 T (•) and 0.25 T (□). The temperature is 125 mK.

layers from five quantities: the slopes and intercepts of the coverage lines, and a measurement of how much ^{3}He was taken out of the cell in going from the higher to the lower coverage. We conclude that the lower line corresponds to one Curie-like, and thus a solid, ^{3}He layer. The upper line corresponds to a coverage of $1\frac{1}{2}$ Curie-like layers and $3\frac{1}{2}$ liquid layers. We note that there is a subtlety in the magnetization of the first liquid ^{3}He layer on a solid ^{3}He monolayer. In particular, both Hammel and Schuhl *et al.* have shown at lower temperatures that about half of the first liquid layer displays Curie-like behavior [1,14]. Finally, the relative amounts of liquid and solid can also be inferred from ^{3}He T_1 (or T_2) versus T measurements using the Hammel-Richardson model [11]. The ^{3}He coverage values inferred from magnetization data, T_1 data, and T_2 data agree to within 5%. The coverage values are also consistent with results obtained from an Argon adsorption isotherm [15].

We now present more details of the magnetization transport results. In an earlier version of the sample and sample cell, Van Keuls observed ^{3}He mediated ^{19}F-^{1}H coupling with a cell full of liquid ^{3}He [9]. This observation led to the current coverage studies. Figure 4 shows the magnetization transport results for three ^{3}He coverages: much less than a monolayer, a solid ^{3}He layer, and a cell full of liquid ^{3}He. We monitor the ^{19}F magnetization as a function of time after a 120° proton pulse. The ^{19}F equilibrium magnetization is 1.0. The data are all taken in a 0.25 T (10.5 MHz) field at 485 mK. The lower curve (also shown in Fig. 1) shows the large amount of magnetization transfer brought about by the existence of a solid ^{3}He layer. We can decouple the substrates, and thus make the effect vanish, by removing the ^{3}He layer. The upper curve shows these data. The decreasing signal is the background suppression due to the data acquisition technique. After the initial proton pulse, we sample the ^{19}F magnetization with a series of 4° ^{19}F tipping pulses. The observed suppression is consistent with the predicted suppression due to this pulse sequence. The full cell data, middle curve, show a smaller cross coupling effect. In terms of the heat transfer analogy, the "film" responsible for the surface transport now has a significant heat capacity. Thus, a change of temperature in one substrate has a smaller effect on the other substrate.

Figure 5 compares the magnetization transport effect at two fields: 0.15 and 0.25 T. The temperature is 125 mK and the ^{3}He coverage is five monolayers. The drop in ^{19}F magnetization is sharper at the smaller field. Since the surface substrate coupling times increase linearly with field [1,2], we also expect the magnetization transport times to increase with field.

The transport effect is temperature independent from 125 to 750 mK. The equilibrium magnetization does change with temperature, but our results are scaled by the equilibrium magnetization. For the solid coverage, this temperature independence can be understood in terms of the temperature independence of the ^{19}F, ^{1}H, and ^{3}He T_1. For the five monolayer coverage, the transport effect could have a temperature dependence due to the T^2 dependence of particle diffusion in the Fermi liquid state. In our experiments, however, Fermi liquid diffusion is fast compared to the surface substrate coupling times and hence is not the rate-determining step. In future experiments, the T^2 dependence might be observable. Our magnetization experiments could then serve as another probe of the Fermi liquid state.

In conclusion, in a system of spatially separated substrate nuclei coupled via a ^{3}He monolayer, we have observed magnetization transfer from one substrate to the other. This transfer is brought about by the combination of spin diffusion, exchange driven ^{3}He substrate coupling, and ^{3}He quantum solid diffusion.

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