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ACCEPTED MANUSCRIPT

Fingerprints of single nuclear spin energy levels using STM - ENDOR

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We performed STM-ENDOR experiments where the intensity of one of the hyperfine components detected in ESR-STM is recorded while an rf power is irradiated into the tunneling junction and its frequency is swept. When the latter frequency is near a nuclear transition a dip in ESR-STM signal is observed. This experiment was performed in three different systems: near surface SiC vacancies where the electron spin is coupled to a next nearest neighbor ^{29}Si nucleus; Cu deposited on Si(111)7x7 surface, where the unpaired electron of the Cu atom is coupled to the Cu nucleus (^{63}Cu, ^{65}Cu) and on Tempo molecules adsorbed on Au(111), where the unpaired electron is coupled to a Nitrogen nucleus (^{14}N). While some of the hyperfine values are unresolved in the ESR-STM data due to linewidth we find that they are accurately determined in the STM-ENDOR data including those from remote nuclei, which are not detected in the ESR-STM spectrum. Furthermore, STM-ENDOR can measure single nuclear Zeeman frequencies, distinguish between isotopes through their different nuclear magnetic moments and detect quadrupole spectra. We also develop and solve a Bloch type equation for the coupled electron-nuclear system that facilitates interpretation of the data. The improved spectral resolution of STM - ENDOR opens many possibilities for nanometric scale chemical analysis.

I. INTRODUCTION

The attempt to detect and manipulate a single spin is a fundamental challenge in nanoscience and nanotechnology. For that purpose, several low temperature scanning tunneling microscopy (STM) techniques have been developed. In particular an electron spin resonance (ESR) detection has been developed by annalyzing the current power spectrum of an STM, a technique known as ESR-STM[1]. A related technique measures DC spin polarized current in presence of variable rf frequency around the Larmor frequency[2,3].

In this work we develop a novel technique for detection of single electron and nuclear resonance. This is based on ENDOR (Electron Nuclear Double Resonance), i.e. a technique where rf field frequencies are swept across nuclear transitions which are then detected via intensity changes of a simultaneously irradiated ESR (Electron Spin Resonance) transition. This is possible when there is a coupling between the electron and the nuclear spins. The spin Hamiltonian is then

$$\mathcal{H}_0 = \gamma_e H_0 S_z + \gamma_n H_0 I_z + \mathbf{S} \cdot \hat{a} \cdot \mathbf{I} \tag{1}$$

where $\mathbf{S}, \mathbf{I}$ are the electron and nuclear spin operators, respectively, γ_e, γ_n are the corresponding gyromagnetic ratios (e.g. for an electron's g factor of 2 $\gamma_e = 2.8$MHz/G and for ^{29}Si nucleus $\gamma_n = -8.4$MHz/T), H_0 is a DC magnetic field in the z direction and $\hat{a}$ is the hyperfine tensor. The electron and nuclear Zeeman energies are defined as $h\nu_e = \gamma_e H_0$, $h\nu_n = \gamma_n H_0$, respectively.

The simplest case $S = I = \frac{1}{2}$ is shown[4] in Fig. 1a. In this case, one has two ESR transitions at $\nu_e \pm \frac{1}{2}a$, where a is the component of $\hat{a}$ parallel to the magnetic field and $a \ll \nu_e$ is assumed, and two nuclear transitions at $|\frac{1}{2}a \pm \nu_n|$. Thus, when $\frac{1}{2}a > \nu_n$ (as in our low field experiments) the two nuclear transitions are separated by $2\nu_n$, identifying the NMR frequencies Fig. 1b. In the usual ENDOR method[5] one of the ESR transitions is saturated so that the level populations become equal and there is no (or little) absorption. Irradiation at the NMR frequency involves a third state with an opposite nuclear spin and therefore will unequilize the ESR levels populations, hence the ESR intensity is partially restored[5,6]. A distinct type of "negative ENDOR"[7–9] is obtained by applying a strong rf field that modifies the ESR signal and then the ESR intensity at the original peak is reduced.

The ability of ENDOR to detect the nuclear transition frequencies, combined with the ability to detect single electron spins by STM techniques, such as ESR-STM, opens the possibility to detect the nuclear transition frequencies of a single atom, once the hyperfine spectrum is detected. This is the topic of this paper. The technique of ESR-STM is capable of detecting single spins. In this method, a Larmor frequency component of the tunneling current is induced by the precession of a nearby single spin on the surface. The existence of this phenomenon has been demonstrated on several spin systems, allowing also observation of hyperfine coupling[1,10–19]. We note that the theoretical understanding of the phenomena seen in ESR-STM is a subject of ongoing research[1,20]. The more recent proposal[20] employs spin-orbit coupling as well as an additional direct current path from the tip to the substrate. Detailed calculations show that the interference between the two paths, i.e. the one via the spin and the direct path, produce a Larmor resonance in the power spectrum of the current, i.e. an ESR-STM effect[20].

In this work we demonstrate for the first time the fea-

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