様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　６ 年　 ６ 月　 ７ 日現在

機関番号：１１３０１
研究種目：基盤研究(C)（一般）
研究期間：2021～2023
課題番号：２１Ｋ０４８７３

研究課題名（和文）Development of Magnetic Microscopy using RF Photon Assisted Tunneling between Superconductor Electrodes

研究課題名（英文）Development of Magnetic Microscopy using RF Photon Assisted Tunneling between Superconductor Electrodes

研究代表者

Ｓｈａｈｅｄ　ＳｙｅｄＭｏｈａｍｍａｄＦａｋｒｕｄｄｉｎ（Shahed, Syed Mohammad Fakruddin）

東北大学・多元物質科学研究所・助教

研究者番号：６０８９９４０９

交付決定額（研究期間全体）：（直接経費）　　3,200,000円

研究成果の概要（和文）：私たちは、サンプル温度が400mKのTOVのラジカル分子を含むSTMトンネリング接合部にRF信号を注入しました。スピン由来のコンド共鳴によりRF由来の特徴を検出しました。私たちは、光子助長トンネリング（PAT）プロセスのTien-Gordonモデルを修正するために、電子-光子結合定数を導入する修正モデルを提案しています。この研究は、RF領域での電子-光子結合定数を調整する経路を示唆しています。別の研究では、TbPc2というSMMにおける交換相互作用エネルギーを制御する新しい方法を見つけました。このエネルギーは、Tb 4fスピンを量子情報処理に使用するための重要なパラメータです。

研究成果の学術的意義や社会的意義
This work may allow for tuning of the electron-photon coupling constant in the RF region which has implications for the future spintronics. We found a way to control the exchange interaction energy in TbPc2 molecule which is crucial for utilizing the Tb 4f spin for quantum information processing.

研究成果の概要（英文）：Combining radio frequency (RF) signals in scanning tunneling microscopy (STM) attracts attention for electron spin resonance detection of a single molecule. We inject an RF signal in the STM tunneling junction, which contains a radical molecule of TOV at the sample temperature of 400 mK. We detected the RF derived signature in the kondo resonance that originated from spin. We propose a model to modify the Tien-Gordon model for the photon-assisted tunneling (PAT) process by introducing the electron-photon coupling constant. This work suggests a pathway to tuning the electron-photon coupling constant in the RF region. In separated study, we found a new method to control the exchange interaction energy in the SMM, TbPc2, which is critical parameter to use the Tb 4f spin for the quantum information process.

研究分野：Surface science

キーワード：Radio frequency　Spin

Background: A combination of scanning tunneling microscopy (STM) with lights attracts wide attention, which is intended to introduce various spectroscopic techniques into the atomic scale space resolution of the STM. Among multiple wavelengths of light, the photon energy of the RF signal covers a spin excitation energy. An example can be seen in recent reports of the electron spin resonance (ESR) using STM set-up, which shows a resonant spin excitation signal for a single atom and molecule whose energy resolution can be compared with the conventional ESR. The dynamic motion of the target spin by the injected RF signal is proposed for a detection mechanism. The phenomenon suggests that the dynamic of the electron and spin is a good target of the investigation.

For a combination of RF and devices, many studies utilized the combination of the single electron transistor (SET); the high frequency of the electric field of the RF signal is used to reveal the dynamics of the electron in the Coulomb blockade region in the frequency region of GHz. The behavior of the many-body effect caused by a coherent motion of the electrons such as a Kondo effect, which is a formation of the spin-singlet state to screen the target spin, is an intriguing target to be investigated with the electric field modulation in the high-frequencyregion. However, though an apparent Kondo effect is observed in SET experiments, a limited number of studies investigated the SET Kondo effect with RF injection.

Purpose of the research: The spectroscopic investigation of the RF injection was started with a conventional superconductor-insulator-superconductor (SIS) junction using a sharp tunneling feature of a superconductor state to investigate the combination with the RF. Meanwhile, the phenomena induced by the RF wave injection in the superconductor (SC) tunneling junction are called photon-assisted tunneling (PATs) and have been studied for a long time. Pioneered by Tien and Gorden,[27] the inelastic tunneling process understands the modification of the tunneling conductance near the superconductor gap. The combination with the RF permits a higher-order tunneling process, a basis of the photon-assisted tunneling (PAT) model. The SIS investigation is now developed into a more versatile STM-based experiment in which more intriguing physical phenomena like multiple Andreev reflections (MARs) are extended to more varied physical systems rather than superconductors.

In this report, we investigate the behavior of the Kondo resonance with the injection of RF signal in the tunneling junction in the STM set-up. Without the RF injection, a Kondo resonance at the Fermi level with 2 meV width is observed by the adsorbate of the TOV molecule, which is a pure organic radical providing an S=1/2 spin state delocalized in the molecule. When an RF signal of 10.4 GHz is injected, the Kondo peak splits into negative and positive peaks. The energy separation of the two peaks becomes more significant proportional to the AC component of the electric field of RF signal at the tunneling junction (V_{ac}) while the integrated intensity is constant. The phenomenon can be accounted for by a modulation model of the tunneling bias modulation with the RF Vac and the PAT model considering multiple phonon adsorption/ejection. However, when RF of 9.0 GHz is injected, we found coexisting of the elastic

Kondo feature (zero-bias peak) and the inelastic peak (split peak), which neither model cannot explain. We propose a model to modify the PAT model of multiple photons by introducing the electron-photon coupling recently presented in a theoretical model. This work suggeststuning the electron-photon coupling constant in the RF frequency range.

Methods: Experimentally, we employed ultra-high vacuum (UHV) STM (Unisoku, Japan), where the sample was cooled using He, and the sample temperature was kept at 400 mK during the entire observation. The introduction of the RF signal to the STM head is explained in the supporting information (SI) but briefly illustrated schematically in Figure 1(a). SubMiniature version A (SMA) cable and connector for RF wave transfer. SMA cables reach the anchor point a few inches separated from the head. We synthesized 1,3,5-triphenyl-6-oxoverdazyl (TOV) molecules following previous recipes. The molecules were sublimed from a heated Ta boat and absorbed on an Au(111) surface at room temperature. TOV is a stable radical whose spin originates from an unpaired p electron at N atom. We previously reported the bonding configuration and the Kondo resonance of TOV molecules adsorbed on the Au(111) surface, executed at the sample temperature of 4.7 K. In the current report, the entire measurement was executed at the sample temperature of 400 mK.

Research findings: We injected an RF signal into the STM tunnel junction containing spin S=1/2 p radical at the sample temperature of 400 mK. The p radical shows a Kondo feature with a width of 2 mV at the Fermi level, which offers a split when an RF signal is injected. The split fans with the power of the RF, which we reproduced with a simulation with two models; one is using the modulation voltage, and the other is assuming a multiple adsorption/ejection of RF photons. However, we observe an elastic component even when the original peak split widely with the RF, which cannot be simulated with both models. We modify the Tien-Gordon model so that the elastic and inelastic components are separated, and the two parts' weight is expressed with a coupling constant λ, which is along a recent theory and can reproduce the experimental result well.

５．主な発表論文等

〔雑誌論文〕　計2件（うち査読付論文　1件／うち国際共著　2件／うちオープンアクセス　0件）

１．著者名 Mohammad Ikram Hossain、Ferdous Ara、Syed Mohammad Fakruddin Shahed、Wang Zhipeng、Komeda Tadahiro	４．巻 14
２．論文標題 Observation of a Yu-Shiba-Rusinov state originating from the magnetic moment in a curved monolayer island of 1T-phase NbSe2	５．発行年 2022年
３．雑誌名 Nanoscale	６．最初と最後の頁 9860～9868
掲載論文のDOI（デジタルオブジェクト識別子） 10.1039/D2NR02007F	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Syed Mohammad Fakruddin Shahed, Ferdous Ara, Mohammad Ikram Hossain, Keiichi Katoh, Masahiro Yamashita, and Tadahiro Komeda	４．巻 XXXX
２．論文標題 Observation of Yu−Shiba−Rusinov States and Inelastic Tunneling Spectroscopy for Intramolecule Magnetic Exchange Interaction Energy of Terbium Phthalocyanine (TbPc) Species Adsorbed on Superconductor NbSe2	５．発行年 2022年
３．雑誌名 ACS NANO	６．最初と最後の頁 XXXXXXX
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acsnano.1c11221	査読の有無 無
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

〔学会発表〕　計5件（うち招待講演　0件／うち国際学会　4件）

１．発表者名 Syed Mohammad Fakruddin Shahed, Mohammad Ikram Hossain, Ferdous Ara, Zhipeng Wang, Tadahiro Komeda
２．発表標題 nvestigation of the spin properties of the TbPc2 (single molecule magnet) molecules and its fragments on the superconductor (NbSe2) surface using STM and STS
３．学会等名 14th International Symposium on Atomic Level Characterizations for New Materials and Devices '22（国際学会）
４．発表年 2022年～2023年

１．発表者名 Syed Mohammad Fakruddin Shahed, Mohammad Ikram Hossain, Ferdous Ara, Zhipeng Wang and T. Komeda
２．発表標題 Superconductor assisted detection of spin in Single molecule magnet (SMM) using scanning tunneling microscopy
３．学会等名 Nano-bio and Advanced Materials Engineering (NAME-2023)（国際学会）
４．発表年 2022年～2023年

１．発表者名
Syed Mohammad Fakruddin Shahed

２．発表標題
Variation of the energy of spin flip excitation depending on the adsorption configuration of terbium phthalocyanine on superconducting substrate adsorbed on superconducting NbSe2.

３．学会等名
JPS 77th Annual Meeting

４．発表年
2021年～2022年

１．発表者名
Syed Mohammad Fakruddin Shahed

２．発表標題
STM-IETS Measurement of Exchange Interaction Energy of Radical Ligand and Metal 4f of SMM TbPc2 on Superconductor NbSe2

３．学会等名
29th International Colloquium on Scanning Probe Microscopy (ICSPM29)（国際学会）

４．発表年
2021年～2022年

１．発表者名
Syed Mohammad Fakruddin Shahed

２．発表標題
Probing the local spin states of the Metal Phthalocyanine (TbPc) molecules and their assembly on superconducting NbSe2 surface

３．学会等名
ALC'21（国際学会）

４．発表年
2021年～2022年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

―

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考
研究分担者	米田　忠弘 (Komeda Tadahiro) (30312234)	東北大学・多元物質科学研究所・教授 (11301)	

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関