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研究課題名（和文）反強磁性体におけるスピン超流動の核磁気共鳴研究

研究課題名（英文）Spin superfluidity in solid antiferromagnets

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研究成果の概要（和文）：強いマイクロ波による励起下では、反強磁性であるMnCo3サンプル内のアンサンブル核スピンの核磁気共鳴は非線形であることを観測した。この実験結果の信号は、核スピンがマイクロ波の励起によって、核スピンの温度が上昇するというような従来のモデルでは説明することが出来ない。実験結果と理論を比べてみると、強いマイクロ波による励起下では、核スピンの磁気モーメントの角度は大きく偏向されていることがわかる(磁気モーメントの強度は変化しない)。この事実はコヒーレントに一様に歳差運動をする核スピンによる核マグノンによるボーズ・アインシュタイン凝縮と同一視することが出来る。

研究成果の概要（英文）：We observed nonlinear nuclear magnetic resonance signals in antiferromagnetic MnCO3 samples at strong microwave pumping. The signals indicated that the conventional model of heating of nuclear spins by microwave pumping can not explain the observed results. We developed a theoretical framework to explain nonlinear nuclear resonance signals at strong microwave pumping. Our approach is based on Landau-Lifshitz-Gilbert equations of motion to describe coupled electron-nuclear spin precession. Comparison of our theory with experiments indicates that under strong microwave pumping the nuclear magnetization vector is deflected by a large angle while preserving its magnitude. This can be identified with the Bose-Einstein condensate of nuclear magnons with coherently and uniformly precesses nuclear spins.

研究分野：condensed-matter physics

キーワード：magnetic resonance

１．研究開始当初の背景

Crystalline ferro- and antiferromagnets with coupled electron-nuclear spin precession have been of interest since 1960-s due to some peculiar properties of the nuclear magnetic resonance (NMR). In particular, the strong hyperfine coupling between electron and nuclear spins introduces an effective interaction between nuclear spins, which results in particular in the dynamical frequency shift of NMR, the so called frequency pulling, and strongly nonlinear NMR signals. Following the seminal work of de Genners (1963), the strongly nonlinear NMR signals in such systems are interpreted in terms of heating of the nuclear spin system by applied RF field.

Recently (Bunkov, 2010), it has been noted that the nonlinear NMR properties of nuclear spins in easy-plane antiferromegnets (e.g. MnCO3, RbMnF3,CsMnF3, etc.) are very similar to those of nuclear spins in superfluid helium-3, where the frequency pulling is also observed. The latter system exhibits a remarkable phenomenon of the Bose-Einstein Condensation (BEC) of nuclear spin waves (magnons). Thus, it was suggested that nonlinear NMR in these crystalline materials should also show signs of the BEC of magnons. This become the main objective of the reported work.

２．研究の目的

(1) The main objective of the proposed research was to investigate the nonlinear NMR signals in MnCO3 easy-plane antiferromagnetic sample and to establish connection with the propped existence of the Bose-Einstein condensate of magnons. Traditionally, the nonlinear NMR signals were interpreted by the heating of the nuclear spin systems, however such an interpretation should give quantitatively different behavior of NMR signals with increasing RF pumping. Such differences could be elucidated by recording both absorption and dispersion nonlinear NMR signals under continuous RF pumping for different powers of RF, and comparing them with the theoretical predication of both heating scenario and scenario of magnon BEC.

(2) In order to make comparison of the experimentally obtained nonlinear NMR signals with the predications of magnon BEC scenario, a proper theory based on Landau-Lifshitz-Gilbert equations of motion for coupled electron-nuclear spin systems needed to be developed. This was another objective of the research.

(3) Finally, it was realized that coupled electron-nuclear precession in easy-plane antiferromagntic material might present a great advantage to study the regime of strong coupling between a single electro-magnetic (EM) field mode in a RF resonator and the nuclear spin system. Usually, the interaction strength between nuclear spins and EM field is too small to reach this regime. However, in the above materials the interaction is mediated by the electron spin system, therefore is strongly enhanced. Observation of the strong coupling regime between EM mode in a resonator and nuclear spin system could be the first such observation.

３．研究の方法

(1) We used conventional continuous-wave (CW) NMR method and both homodyne and heterodyne detection techniques to measure both absorption and dispersion nonlinear NMR signals in an easy-plane weakly-ferromagnetic antiferromagnet MnCO3 sample. We have built a several setups based on the above methods, employing both Schottky-type RF detector for homodyne detection and RF mixers for heterodyne detectors. For the later experiments, we also employed a commercial network analyzer.

(2) In order to enhance sensitivity of our detection scheme, as well as study the regime of strong coupling between EM mode and nuclear spin system, we developed and fabricated a single-mode

high-quality-factor (~1,000-10,000) spit-ring resonators at frequencies around 600 MHz suitable to study nonlinear NMR signals in MnCO3.
(3) Finally, we developed and fabricated a home-made superconducting magnet capable reaching magnetic fields up to 1 Tesla. In later experiments, we also employed a commercial 9 tesla superconducting magnet from American Magnetics.

４．研究成果

(1) In the first experiments, we developed a relatively high-Q factor (~1,000) single mode split-ring RF resonator and employed a simple detection scheme using a room-temperature Schottky-type RF detector to measure reflected signal from the resonator loaded with the MnCO3 sample. The loaded resonator was cooled down to temperature about 1 K and the NMR frequency of nuclear spins in the sample was tuned to the frequency of resonator by applying the magnetic field of several hundreds of kilo-Gauss. We used a low RF power to stay in the linear regime of NMR. We observed the splitting in the cavity reflection spectrum, which is a characteristic feature (kind of Rabi-splitting) of the strong coupling between the resonator mode and the nuclear spin ensemble in MnCO3. This is, to the best of our knowledge, is the first observation of the strong coupling regime in the nuclear spin ensembles.
The observed spitting in the resonator reflection spectrum showed characteristic scaling as root of the number of spins, as expected from the theory. From analysis of data we obtained cooperativity factor C=0.2 which is close to the condition of strong coupling. In subsequent experiments we employed a higher-quality factor (~10,000) resonators to obtain cooperativity factor larger than 1, thus clearly demonstrating the strong coupling regime.
While this observation was not our main objective, it represents an interesting finding with possible applications for quantum technologies. Recently, there has been a significant interest to the strong coupling of ensembles of quantum particles to the resonator modes due to possible applications for quantum memories, quantum transducers, quantum hybrid systems, etc. The nuclear spins can provide additional advantages of longer decoherence time comparing with the electron spins. Further work on nuclear ensembles strongly coupled to RF resonators is promising.
(2) In the following experiments we concentrated on the study of nonlinear NMR signals at high RF powers. To avoid complications arising from the strong coupling of spin system to the resonator mode, we conducted these experiments in the dispersive regime at NMR frequencies far from the resonator frequency. To obtain both amplitude and phase of the nuclear spin system response, we employed homodyne and heterodyne detection spin to separately detect the absorption and dispersion signals. Both signals showed characteristics features of the nonlinear NMR with frequency pulling, such as frequency shift and hysteretic bahaviour with increasing RF pumping. More importantly, very large signals at high powers of RF pumping indicated that the magnitude of the nuclear magnetization vector does not decrease with RF power, as would be expected from the conventional heating scenario. Thus, an alternative explanation of the observed NMR signals was required.
(3) In order to explain our experimental results, in collaboration with theoreticians we developed a theoretical model of nonlinear NMR in coupled electron-nuclear spin system based on the Landau-Lifshitz-Gilbert equations as opposed to the conventional Bloch approach. In our theory, the magnitude of the nuclear magnetization vector is

preserved owing to extremely large (order of 600 kilo-Gauss) hyperfine field experienced by the nuclear spins. Using our theory, we could fit the observed experimental signals and show that the nuclear magnetization vector is deflected by angles as large as 70 degrees at highest RF power. This corresponds to the uniform precession of nuclear spins, thus can be identified with the BEC of nuclear spin waves with k=0. This can be stated as the main result of the research.

５．主な発表論文等
（研究代表者、研究分担者及び連携研究者には下線）

〔雑誌論文〕（計２件）

(1) L. V. Abdurakhimov L. V., Bunkov Yu. M., Konstantinov D. "Normal-mode splitting in the coupled system of hybridized nuclear magnons and microwave photons", Phys. Rev. Lett. 114, 2015, 226402 DOI:10.1103/PhysRevLett.114.226402 (2015)
(2) Abdurakhimov L. V. , Borich M. A., Bunkov Yu. M., Gazizulin R. R., Konstantinov D., Kurkin M. I., Tankeyev A. P. "Nonlinear NMR and magnon BEC in antiferromagnetic materials with coupling between electron and nuclear spin precession", Phys. Rev. B 97, 2018, 024425 DOI:https://doi.org/10.1103/PhysRevB.97.024425

〔学会発表〕（計３件）

(1) Abdurakhimov, L. V. "Strong coupling between hybridized nuclear magnons and microwave photons", invited seminar at TU Delft, the Netherlands, June 2015
(2) Konstantinov, D. Normal-mode splitting in the coupled system of hybridized nuclear magnons and microwave photons", invited seminar at University Paris-Sud, France, March 2015.
(3) Abdurakhimov, L. V. "Strong Coupling of Hybrid Nuclear-Electron Magnons to a Microwave Resonator", contributed talk at Magnetism 2015, Sheffield, United Kingdom, April 2016

〔図書〕（計０件）

〔産業財産権〕

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〔その他〕
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６．研究組織

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