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研究課題名（和文）僅かな分子の構造変調が劇的なスピン状態変化をもたらす材料群の研究

研究課題名（英文）Study on magnetic materials showing drastic spin state transition upon relatively small structural transformation

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研究成果の概要（和文）：分子磁性という学問分野は我国が世界に誇れる基幹物理化学の一つである。この分野発の新規材料群で、学界・産業界に貢献することは重要である。有機材料の柔軟性に起因して、ラジカルと遷移金属イオンからなる物質の中からスピン転移と構造転移がカップルする系をいくつか見いだした。(1) 有機ラジカルだけからなる結晶性固体で、重合・解重合によるスピン消失・出現が見出された。(2) ラジカルと遷移金属イオンからなる系から、スピン転移材料の２例を得ることができた。これはスピンクロスオーバーの機構として全く斬新なものであった。

研究成果の学術的意義や社会的意義
分子磁性という学問分野は我国が世界に誇れる基幹物理化学の一つである。この分野発の新規材料群で、学界・産業界に貢献することは重要である。本研究推進においては、Dalton Trans. 誌の内表紙採用(2016)、Bull. Chem. Soc.Jpn. 誌のBCSJ Award受賞と表紙採用(2016)、Inorg. Chem. 誌で Editors' Choice (2017)、New J. Chem. 誌で裏表紙採用(2019)、および印刷中ながら Org. Lett. 誌で内表紙採用(2019) などのように、外部評価も概して高い。学術上の意義を反映していると考えられる。

研究成果の概要（英文）：Solid-state magnetic switches are of increasing interest for future application to sensing, memory, display, etc. The authors and co-workers have developed unconventional spin transition/crossover materials, where the sense of organic chemistry has competently been adopted. The first example is a supramolecular chemistry of genuine organic nitroxide biradicals with a triplet ground state, which undergo stepwise polymerization/depolymerization in a crystal. The second example belongs to 2p-3d heterospin systems where the nitroxide-nickel(II) or -copper(II) bond rotates and switches exchange coupling. As these examples show, single-crystal-to-single-crystal structural transitions are often recorded, thanks to small atomic dislocation. Molecular motion in crystalline solids is observed more frequently than expected. Attentive crystallographic study affords sure proof for the spin transition phenomenon.

研究分野：有機材料科学

キーワード：分子性磁性体　集積型金属錯体　超分子科学　単分子磁石　高スピン分子　相転移

様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９、ＣＫ－１９（共通）

１．研究開始当初の背景

分子磁性という学問分野は我国が世界に誇れる基幹物理化学の一つである。この分野発の新規材料群で、学界・産業界に貢献することは重要である。一方で、分子性材料が無機物質では考えられなかったような特性も有することから、「分子性物質ならでは」という性質の導入も必要である。特に分子骨格の柔軟性を本研究課題の鍵とする。我々はこれまでに有機無機の区別なく、4f, 3d, 2p スピンを組み合わせた錯化合物で研究を進めてきた。本課題ではこのような複合スピン系に、柔軟性、加工性、反応性、光吸収などの有機由来・分子由来の機能を導入し具体化する。

２．研究の目的

具体的には、環境応答型磁石、可溶化磁石、光応答性電導体、誘電性磁性体、電導性磁性体を目指した合成開発を目指す。物性物理側からのアプローチとして、極低温、高磁場の研究者とも協力して新素材の物性解析も進めた。ヘテロスピン系に期待されるスピン間交換相互作用については、その相互作用が幾何的配置に大きく左右されるため、構造磁性相関図の提案も興味深い。

３．研究の方法

メンバーは申請代表者と、研究協力者主席研究員小金民造博士、連携研究者岡澤厚助教（東大）、金友拓哉博士研究員（東京理科大）、および電通大基盤理工学専攻の学生のべ十数名である。分担業務として、物質の合成開発、物性測定、理論解析を行った。各種分光法による同定、結晶構造解析も自前で行った。

４．研究成果

有機材料の柔軟性に起因して、ラジカルと遷移金属イオンからなる物質や純粋な有機材料の中からスピン転移と構造転移がカップルする系を多く見いだした。

(1) ニッケル-ビスニトロキシド錯体においてスピン転移材料を得ることができた。非常に緩やかなスピン転移を伴い、単結晶-単結晶構造相転移を見せた。この転移の最中に単結晶構造解析を行い、コマ撮りムービーを作成することもできた。なお、$S = 2$ と $S = 0$ をスイッチする材料は、鉄(II) 錯体以外では初めての例であり、これはスピン転移の機構として全く斬新なものであった。

(2) 鉄(II)イオンを用いたスピンクロスオーバー（SCO）錯体において pyridine 環の 4 位に置換基を導入し配位子場を変化させ SCO 温度を制御することができた。SCO 温度の置換基依存性をハメット定数を用いて議論した。SCO 温度は分子軌道計算（DFT）から予言できることも示した。

(3) 複合機能性を求めて､磁気交換カップリングを有するコバルト(II)イオンを用いた SCO 錯体を開発した。この物質は低温側で １ボーア磁子, 高温側で５ボーア磁子, 途中の温度領域では３ボーア磁子を示す温度依存スピン転移材料であることがわかった。

(4) 零次元磁石にはナノサイズ材料の開発という重要な意義がある。希土類-有機ラジカル直接配位系では配位原子自体をスピン中心にすることができるから、交換相互作用は強くなる。ところで、Gd-ラジカル間には強磁性的・反強磁性的カップリングの双方が観測されているものの、このような系で配位構造との関連はこれまで論じられていなかった。我々は，この系で構造磁性相関を提案し、その相関図に基づく分子設計から、史上最強の強磁性的カップリングおよび反強磁性的カップリングを有する系をそれぞれ開発することができた。さらにこれを単分子磁石開発の設計指針に応用した。

５．主な発表論文等

〔雑誌論文（すべて査読有り）〕（計 51 件）

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〔その他〕
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