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Title:	Experimental Investigation of Magnetism in Natural silk Polymers
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Abstract:	<p>Over 400 million years of evolution, Nature has perfected the structure of silks secreted by arthropods to support various aspects of their life. Extensive research has discovered that the molecular structure of silks has been engineered to encapsulate many remarkable mechanical, optical, and biological properties surpassing even artificial materials. Surprisingly, these insects produce multi-functional silk biopolymers using elementary processing of simple amino acids in water. Materials like steel and Kevlar require much more complex manufacturing processes but still fail to match (spider and silkworm) silk's toughness, albeit their mechanical strength is equivalent to silk's. In short, natural silks are a model material for the future and a fine example of how much Nature has to offer humanity if we look closely enough. A large part of recent research is focused on mimicking silks to produce advanced materials for biomedical and technological applications as they offer eco-friendly, biocompatible, and sustainable alternatives to existing solutions. In the present work, we discuss the intrinsic magnetic properties of natural silk polymers, namely spider dragline silks and mulberry silkworm silks. We present a thorough magnetic study of silks, like their magnetic behavior, the possible origin of magnetism, and magnetic ordering. We discovered silk biopolymers act as amorphous magnets with a stable, sustainable ferromagnetic character from very low (5 K) to high temperatures (400 K). Notably, the magnetization of spider dragline silks is more significant than most organic magnets. Investigation into the source of magnetism revealed that the magnetism possibly originates due to the presence of persistent organic radicals in natural silks. Therefore, silks offer a new novel route to fabricate stable organic magnets with exceptional strength and toughness unmatched by the presently known ones. We established our claims by experimenting on more than 20 silk samples obtained from multiple spiders (Araneus and Neoscona families) and silkworms (Bombyx mori) using SQUID magnetometry, EPR spectroscopy, and various elemental analysis techniques like EDX, XPS, XAS, and ICPMS. These are the first reports of magnetic experiments to show that atomic defects in the building blocks of pure silk fibers induce significant permanent magnetization, thereby opening their direct magnetic applications. Atomistic defects in silks result in the formation of radicals in them. These radicals are sterically protected in silk's structure up to very high temperatures. Radicals in close vicinity tend to interact with each other directly. These interactions can be ferromagnetic or anti-ferromagnetic, depending on the configuration and distance of neighboring radicals. The interacting radicals form small clusters trapped in the glassy state in the silk protein matrix and are distributed throughout the silk's structure, leading to an overall magnetic response. We showed that one could externally control the magnetization of silks by manifesting additional defects (or deformations) in their structure using simple techniques like stretching and cutting. Moreover, during the thermomagnetic analysis, we discovered a unique magnetic transition in all silk samples around 120 K, irrespective of their host insect, i.e., spiders or silkworms. Using a sensitive torsion pendulum that mimicked a spider suspending from its dragline silk, we discovered that even a micrometer-thin silk thread of spider dragline silk could sense magnetic fields up to 30 μT in ambient conditions. We demonstrate that silks can act as bio-magnetosensors. Moreover, we also developed unique silk-based tiny swimmers/propellers that act as organic magnetic robots (Bio-Magbots). These silk-based bio-magbots can be controlled externally under small magnetic fields to manipulate microscopic organic and inorganic matter. We explicitly illustrated that silk-based magnetic robots could perform the non-contact on-demand translation and rotatory motions. Silk-based magnetic robots could also magnetically trap organic and inorganic material at a specific location using small localized magnetic fields (\approx 20 mT). In a remote-controlled fashion, we demonstrated the transport of various cargo, such as plant tissues, animal tissues, polymer bundles, and non-magnetic immiscible chemicals, on the surface of the water using silk swimmers. Silk-based bio-magbots can perform targeted drug delivery, magnetic trapping, and bioremediation in a cell-like liquid environment. Furthermore, magnetically actuated artificial muscles and scaffolds for tissue engineering could be developed using silk and silk-based materials. Our work also opens the intriguing potential of silk in developing ultra-lightweight and thin bio-magnetosensors and nanoelectromechanical systems (NEMS) made from sustainable materials. The present research establishes natural silk polymers as amorphous magnets and opens a new route to developing organic magnets by biomimicking silk proteins. Unlike most organic magnets, silks can sustain their ferromagnetism for a very long time (years) without any decrease in their magnetization. They can be easily stored at room temperature and 50% relative humidity. Natural silks can be an archetypal polymer for developing organic magnets that sustain ferromagnetism up to high temperatures and simultaneously have silk's excellent physical properties (mechanical, optical, etc.). Such protein-based magnets will have a unique advantage in biomedical sectors. The work has implications in material science, materiomics, quantum biology, biomedical engineering, and mechanical engineering.</p>
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