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Perspective

Important Elements of Spin-Exciton and Magnon-Exciton Coupling

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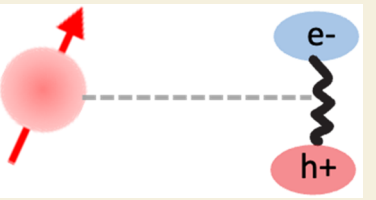
ABSTRACT: The recent discovery of spin-exciton and magnon-exciton coupling in a layered antiferromagnetic semiconductor, CrSBr, is both fundamentally intriguing and technologically significant. This discovery unveils a unique capability to optically access and manipulate spin information using excitons, opening doors to applications in quantum interconnects, quantum photonics, and opto-spintronics. Despite their remarkable potential, materials exhibiting spin-exciton and magnon-exciton coupling remain limited. To broaden the library of such materials, we explore key parameters for achieving and tuning spin-exciton and magnon-exciton couplings. We begin by examining the mechanisms of couplings in CrSBr and drawing comparisons with other recently identified two-dimensional magnetic semiconductors. Furthermore, we propose various promising scenarios for spin-exciton coupling, laying the groundwork for future research endeavors.



KEYWORDS: *magnetic semiconductor, exciton, magnon, spin-exciton coupling, magnon-exciton coupling*

INTRODUCTION

Historically, magnetic semiconductors have been of interest in information technology, where their semiconducting and magnetic properties are particularly useful for logic and information storage, respectively. Two distinct classes of magnetic semiconductors have been explored in this context: intrinsic magnetic semiconductors, which contain dense lattices of magnetic ions, and dilute magnetic semiconductors, in which a small percentage of magnetic ions are incorporated into nonmagnetic semiconductors. Among these two classes, dilute magnetic semiconductors have generally been the focus of applied research, as the ability to utilize existing fabrication processes for developed semiconductors facilitates their integration into devices.[1,2] To this end, research surrounding magnetic semiconductors has focused on attaining magnetic ordering temperatures above room temperature while maintaining high carrier mobility. In recent years, however, increased interest in using these magnetic semiconductors for applications in quantum information science, quantum photonics, and opto-spintronics has shifted the research focus toward intrinsic magnetic semiconductors.[3−5] These emerging fields demand materials possessing magnetic moments highly uniform from those attainable with dilute magnetic semiconductors.

The potential of intrinsic magnetic semiconductors in the context of these quantum technologies is derived from the coexistence of magnons and excitons. Magnons are important because they can be directly or indirectly coupled to qubits.[6,7] For instance, magnons that hold magnetic dipole moments can directly couple to spin-based qubits through magnetic dipolar coupling.[8] In addition, magnons can be also indirectly coupled to superconducting qubits when placed in a microwave cavity.[9] Bright excitons, on the other hand, play crucial roles in optoelectronics by facilitating the interaction, emission, and control of light in electronic devices. Excitons are important elements in photodetectors, optical switches, and lasers.[10] Extensive research efforts have been devoted to investigating exciton physics and its applications in monolayers of two-dimensional (2D) transition metal dichalcogenides (TMDCs). These materials have excitons with large oscillator strength confined within an atomically thin layer.[11] They hold the potential to shrink the device size down to the atomic layer and to tune the device property by forming various heterostructures.[12]

While magnons and excitons individually offer valuable properties, coupling between the two in a magnetic semiconductor could give rise to more exotic phenomena and diverse device applications. As one example, by creating and manipulating excitons with light, it would be possible to read out or control magnons that are coupled to qubits. In addition, excitons that absorb and emit light can be manipulated by magnons, introducing a spin degree of freedom in optoelectronics. The coupling of magnons and excitons also permits interesting opportunities to manipulate real magnetic moments using various photonics platforms.

Despite the obvious technological appeal of such exciton−magnon coupling, materials displaying these phenomena have largely remained elusive. However, this coupling was recently discovered in the 2D magnetic semiconductor, CrSBr.[13,14] In

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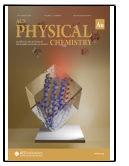

this Perspective, we aim to address these questions: What is the origin of exciton−magnon coupling in CrSBr? Why is a similar coupling not observed in other 2D (or 3D) magnetic semiconductors? What design principles can be implemented in new materials to improve the coupling found in CrSBr? The objective of this review is to help readers understand the spin-exciton coupling in CrSBr and to motivate and guide synthetic efforts from material scientists to broaden the library of this class of materials.

The structure of the paper is as follows: first, we discuss the details behind spin-exciton and magnon-exciton coupling in CrSBr; second, we compare CrSBr to other magnetic semiconductors; and lastly, we explore exciting ways to tune and modify the existing spin-exciton coupling.

ELECTRONIC BAND AND MAGNETIC STRUCTURE OF CrSBr

In order to understand excitonic properties in CrSBr, it is first crucial to understand its electronic band structure and magnetic structure. Many of the details of the electronic and magnetic properties of CrSBr can be linked to its local crystal structure; as such, we begin with a brief analysis of the crystal field for Cr in this structure. CrSBr is a member of the FeOCl structural family, where each van der Waals layer consists of a Cr/S double layer capped on each side by bromide ions. The first coordination sphere for each Cr ion is a $CrBr_2S_4$ distorted octahedron with C_{2v} point symmetry. By comparison to an ideal octahedron (Figure 1a), the primary distortion is a bend

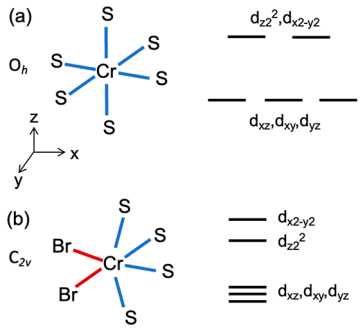

Figure 1. (a) CrS_6 and (b) CrSBr distorted octahedra with their respective crystal field splitting.

in the S−Cr−S bond angle oriented along the *b*-axis (Figure 1b). This structural distortion should stabilize a σ^* antibonding orbital with orbital density primarily oriented along the crystallographic *b* axis (labeled d_{z^2} in Figure 1). For the $3d^3$ electron configuration of Cr^{III} ions in CrSBr, this stabilized orbital represents the lowest unoccupied molecular orbital (LUMO), such that the conduction band of CrSBr should be derived from orbitals that extend primarily along the *b* axis.

Such chemical intuition obtained from the crystal field splitting can be directly compared with the calculated electronic band structure of CrSBr. The electronic band structure of CrSBr is calculated using first-principles within the *ab initio* GW framework[13] and the atomic orbitals contributing to these bands are identified.[15] The orbitals contributing to the valence band minima (VBM) are mixtures of Cr $3d_{xy}$, $3d_{yz}$, and $3d_{xz}$ and Br $4p_z$ orbitals, and those contributing to the lower branch of conduction band minima (CBM) are Cr d_{z^2}, $d_{x^2-y^2}$ and S p_z,[15] which is in good agreement with the simple orbital model in Figure 1. Additionally, the CB is highly anisotropic, with little dispersion along Γ−X (corresponding to the crystallographic *a* axis) but substantial dispersion along Γ−Y (corresponding to the crystallographic *b* axis).[13] This band structure is suggestive of quasi-1D behavior with high mobility only along the *b* axis, which is again consistent with our crystal field model. An important consequence of this covalent bonding along the *b* axis is that the spatial delocalization of the electron density of excitons in CrSBr extends over 20-unit cells along the *b* axis while they are less delocalized along the *a* axis, in about a 2:1 ratio.[13]

The magnetic structure of CrSBr can similarly be explained by an examination of the local crystal structure. For a pseudo-octahedral Cr^{III} ion, the Goodenough−Kanamori−Anderson rules predict ferromagnetic (FM) exchange interactions for bond angles near 90° and antiferromagnetic (AFM) interactions for bond angles near 180°.[16] Similarly, along the *c* axis, there is a competition where AFM interactions slightly outweigh FM interactions.[17] The magnetic exchange interactions in CrSBr are dominated by its three nearest-neighbor superexchange pathways. For the exchange interactions along the *a* axis and diagonal across the *ab* plane (J_2 and J_1, respectively), Cr ions are bridged by S or Br ions with bond angles near 90°, which should generate FM exchange interactions. Along the *b* axis, Cr ions are bridged by S ions with bond angles of 162°, which should generate competing AFM and FM exchange interactions (J_3). Consistent with this structural analysis, inelastic neutron scattering measurements reveal that all three superexchange pathways are FM, and that the magnitude of J_3 is the smallest.[18] These three FM superexchange pathways lead to in-plane ferromagnetism for CrSBr. While much weaker than the in-plane exchange interactions, the interlayer exchange coupling in CrSBr is AFM. Collectively, this induces A-type AFM order in CrSBr, as depicted in Figure 2a. This magnetic structure causes the VB

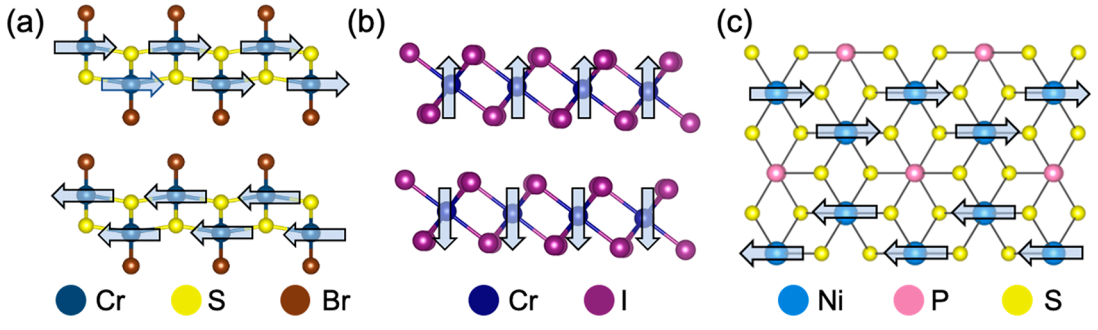

Figure 2. Crystal and pin structure of the systems discussed in the paper. (a) CrSBr, (b) CrI_3, and (c) $NiPS_3$.

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and CB for each layer of CrSBr to be spin-polarized but with opposite spin polarizations for adjacent layers. Triaxial magnetocrystalline anisotropy in CrSBr causes spins to orient in-plane along the crystallographic *b* axis (the magnetic easy axis).

The van der Waals structure of CrSBr generates intralayer and interlayer exchange interactions separated by several orders of magnitude. While the intralayer spins are coupled ferromagnetically with a spin exchange interaction in the range of 1 meV,[18,19] the antiferromagnetic interlayer spin exchange interactions are orders of magnitude weaker (10 ueV).[14] Consequently, the magnon energy of CrSBr falls within the GHz range, primarily due to the small interlayer AFM exchange interactions in comparison with other terahertz AFM magnons.

SPIN-EXCITON AND MAGNON-EXCITON COUPLING IN CrSBr

Having established the individual properties of excitons and magnons in CrSBr, we now turn to a discussion of their coupling. The electronic band structure of a monolayer of CrSBr reveals that both the CB and VB exhibit majority spin polarization. Consequently, excitons maintain this spin polarization upon photoexcitation. In the case of bilayer CrSBr in the AFM ground state, where the two layers have antiparallel spin configurations, individual excitons are confined to a single van der Waals layer with specific spin polarization. Electronic hybridization between the valence and conduction bands of the two van der Waals layers is spin-forbidden, which prevents excitons from delocalizing to the adjacent layer. In the presence of a sufficiently large applied magnetic field, spins in both layers become polarized in the same direction such that interlayer hybridization is spin-allowed, and electrons and holes can delocalize across both layers. In this context, the degree of hopping is determined by the spatial and spinor parts of exciton wave function overlap between layers.[13] Since the interlayer spacing is fixed and applying an external magnetic field does not induce any structural change, the degree of delocalization tracks with the parallel component of spins. Treating spin-exciton coupling as a second-order perturbation to GW-BSE exciton energy calculation, the calculated energy difference between FM and AFM excitons is 20 meV, which is in excellent agreement with the experimentally measured exciton energy difference from the field-dependent steady-state photoluminescence and reflectance.[13]

This spin-exciton coupling is a critical element, leading to exciton-magnon coupling in CrSBr. Upon optically launching coherent magnons, it creates a dynamic modulation of the spin canting angles between layers. This means that the spin parallel component, which dictates the electron delocalization of the exciton, gets modulated at the magnon frequency. As a result, when we employ time-resolved optical spectroscopy, we can directly measure coherent magnon response by temporally tracking exciton energy change.[14] In this measurement, transient reflectance spectra contain coherent oscillations across all wavelengths of excitons, and the frequency of these optically measured oscillations (24 and 34 GHz) are in excellent agreement with magnon frequencies in CrSBr, separately measured from magnetic resonance spectroscopy (22 and 37 GHz).[20] Interestingly, there is a 180° phase flip in the coherent oscillations at the exciton resonance peaks. While the multiple exciton peaks stem from the formation of self-hybridized exciton-polaritons,[21,22] the phase flip at the exciton resonance peaks indicates the coherent modulation of the exciton energy. This result strongly supports exciton-magnon coupling in CrSBr and the consequence of this coupling is that magnons in 0.1 meV range can modulate exciton energy by 4 meV.[14] Additionally, coherent magnon transport is also investigated,[23,24] where the dipolar interactions govern the spatial propagation due to the weak AFM interactions.

COMPARISON OF CrSBr TO OTHER 2D MAGNETS

From the mechanism discussed above, it is apparent that the coupling of spin-exciton and magnon-exciton in CrSBr arises from spin-dependent electron and hole hopping linked to its A-type AFM structure.[13] A natural question is whether such a mechanism should extend to all A-type antiferromagnets or, perhaps more broadly, to all van der Waals antiferromagnets. In this section, we examine the absence of exciton−magnon coupling in two other 2D antiferromagnets—CrI_3 (with an A-type AFM structure) and $NiPS_3$ (with in-plane, stripe AFM order)—and discuss what distinguishing properties of CrSBr may enable this coupling.

To begin, we distinguish the types of spin-exciton interactions observed in CrI_3 and $NiPS_3$. Conducting the same magnetic-field dependent measurement of the exciton energy, minimal to no change in exciton energy is observed in CrI_3 and $NiPS_3$.[25−30] However, it is noteworthy that excitons in both CrI_3 and $NiPS_3$ are highly polarized, and the polarization of PL emission direction depends on their spin structures. For example, in $NiPS_3$, exciton emission is only observed when magnetic order is established, and the emission polarization is perpendicular to the zigzag AFM spin directions (Figure 2c).[30,31] In the case of CrI_3, exciton emission displays helical polarization, stemming from out-of-plane uniaxial spin anisotropy (Figure 2b).[29] Despite both CrI_3 and $NiPS_3$ excitons being spin-dependent, the exciton energy remains independent of magnetic ordering. This suggests the absence of a similar degree of electron and hole hopping in CrI_3 and $NiPS_3$, even when spins transition from AFM to FM, compared to CrSBr. One important thing to note is that compared to other 2D magnets, CrSBr has a significantly larger oscillator strength, which is a few times greater than that in TMDCs.

A key distinction between CrSBr and other magnetic semiconductors lies in the size of its excitons. The magnetic semiconductor $NiPS_3$ and the magnetic insulators CrI_3, have spatially localized, Frenkel-type excitons.[26,32,33] The real space electron and hole density calculation of four different excitons of CrI_3 show that electron density extends over 1−3 Cr atoms[32] and the experimentally extrapolated exciton size of $NiPS_3$ is 0.6 nm.[26] From a chemical perspective, the two main factors in achieving covalent bonding and substantial band dispersion in a semiconductor are a small energy difference between metal and ligand orbitals and a correct geometry for orbital hybridization. For CrSBr, σ-symmetry orbital overlap between Cr 3d and S 3p orbitals fulfills this condition, leading to substantial band dispersion along the Γ−Y direction. In contrast, weaker orbital overlap ($NiPS_3$) and large metal−ligand energy differences (CrI_3) lead to flat bands in many other 2D magnetic semiconductors[30,34] We hypothesize that the spatially delocalized exciton in CrSBr plays a significant role in creating spin-dependent electron delocalization, suggesting that high charge mobilities may be necessary to achieve exciton−magnon coupling. In this regard, the recently studied, $CrPS_4$ could be a promising magnetic semiconductor with the desired spin-exciton coupling.[35] In short, we infer that

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CrI_3 and $NiPS_3$ have large exciton binding energies based on their small exciton size, which leads to a lack of electron and hole hopping.

DIFFERENT SCENARIOS OF SPIN-EXCITON AND MAGNON-EXCITON COUPLING IN CrSBr

We now established that spin-dependent electron and hole hopping can modify spatially delocalized excitons in AFM magnetic semiconductors. Looking forward, materials design should focus not just on identifying new materials hosting spin−exciton and exciton-magnon coupling but specifically on finding materials that improve upon the properties already observed in CrSBr. Here, we explore three distinct approaches to enhance these couplings or to observe new physical phenomena derived from these couplings (Figure 3).

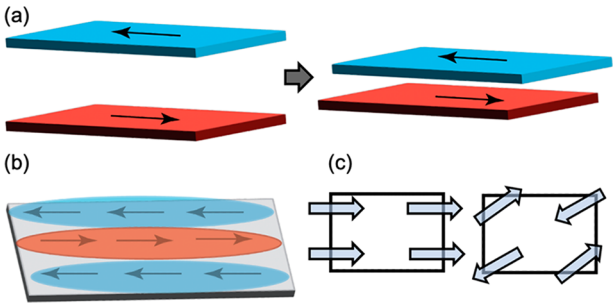

Figure 3. Proposed ways of tuning spin-exciton coupling by (a) decreasing the distance between two spin polarized layers, (b) creating an intralayer AFM spin chain, and (c) changing collinear to noncollinear spin configurations.

One potential limitation in CrSBr is the relatively small shift in the exciton energy induced by the change in the magnetic structure from AFM to FM. Given that CrSBr is a van der Waals material, this energy change is likely linked to the weak orbital overlap between van der Waals layers, which limits the strength of interlayer hybridization. In line with this expectation, a recent study demonstrated that hydrostatic compression of CrSBr greatly reduces the interlayer spacing and, in turn, enhances the spin-exciton coupling (Figure 3a). Under the same magnitude of the applied magnetic field, a more substantial change in the exciton energy change of 60 meV was observed, indicating a 3-fold enhancement of the spin-exciton coupling.[36] Chemical routes to enhance this coupling in CrSBr at ambient pressure are, therefore, of immediate interest, and more generally, this result may indicate that layered, non-van der Waals magnetic semiconductors with stronger interlayer coupling may be desirable synthetic targets.

An alternative approach to strengthen this hybridization is to find materials with *intra*layer spin-dependent excitons, which requires intralayer spin exchange interactions to be AFM in at least one direction (Figure 3b). To explain the effect of such a change, we consider a hypothetical material with the structure of CrSBr, but with stripe AFM order (i.e., FM interactions along one in-plane direction, but AFM along the other). Here, this hypothetical material resembles the magnetic and electronic structure of $NiPS_3$ and CrSBr, respectively. For this magnetic structure, the strength of interchain hybridization and thus the strength of spin-exciton coupling might be orders of magnitude larger than the interlayer hybridization observed experimentally in CrSBr. Such a scenario would be particularly interesting if the coupling strength could approach the exciton energy. Although the spin-exciton coupling in CrSBr operates in the perturbative regime, once the coupling strength becomes similar to the energy of exciton, then it would belong in the ultrastrong regime, where a new ground state could emerge.[37] As an additional benefit, intralayer AFM interactions would push magnons into the AFM magnon transport regime, yielding highly dispersive magnon bands and extremely high group velocities.[38] We note, however, that such THz magnons would require energy down conversion to couple to qubits operating at GHz frequencies. Thus, finding materials with magnons at both GHz and THz frequencies remains a point of interest.

Lastly, we discuss the more exotic scenario in which exciton−magnon coupling could be achieved in a material with a noncollinear spin structure (Figure 3c). All of the materials discussed here, including CrSBr, host collinear magnetic structures in which all of the spins are parallel to each other. In materials with strong magnetic frustration, spin−orbit coupling, or broken inversion symmetry, magnetic interactions such as Dzyaloshinskii−Moriya (DM)[39,40] interactions can promote noncollinear magnetic structures, including topological spin textures. An interesting question then is, if a similar type of spin-exciton coupling can coexist with these spin textures, and if so, might this be a route to generate exciton textures, or to control spin textures with light? To this end, the van der Waals character of CrSBr presents a potential path forward. A van der Waals assembly of multilayer CrSBr with identical twist angles between each layer may create a chiral spin structure along the stacking axis, which could be probed by studying the exciton behavior. Relatedly, recent neutron scattering work in CrSBr has discussed the potential emergence of a topological magnon state if the DM interaction could be enhanced, for example, by assembly with a second material hosting strong spin−orbit coupling.[18] Additionally, increased DM interactions allow skyrmion formation[41] and we may be able to optically read out and manipulate skyrmionics dynamics using spin-exciton coupling.

CONCLUSION

In summary, CrSBr stands out as a unique type of 2D magnetic semiconductor featuring spatially delocalized excitons, similar to those excitons in monolayer TMDCs, while possessing magnetic ordering down to the monolayer. These characteristics open up exciting opportunities to merge optoelectronic properties investigated with excitons in TMDCs and spintronics studied with conventional magnetic materials. The presence of spin-exciton coupling permits particularly interesting opportunities in quantum information science and quantum photonics, where exciton-coupled magnons can shuttle qubit information in a manner that is optically accessible. We provide interesting directions to tune spin-exciton coupling that ranges from enhancing the spatial part of the exciton wave function to creating chiral spin textures. We anticipate numerous creative approaches to build upon this initial discovery. The ongoing extensive research efforts in CrSBr are poised to establish fundamental design principles for tailoring tunable spin-exciton coupling in various magnetic semiconductors. These advancements will take crucial roles in the fields of quantum information science, quantum photonics, and opto-spintronics.

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Notes

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