

Role of photonic interference in exciton-mediated magneto-optic responses

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(Received 9 March 2026; revised 4 June 2026; accepted 13 July 2026; published 3 August 2026)

Coupled optical and magnetic excitations can give rise to remarkably strong magneto-optic responses. This is particularly evident in van der Waals magnets, such as the antiferromagnet CrSBr, where excitons and magnons emerge from the same electronic orbitals. While previous work has primarily focused on uncovering the origin of the resulting exciton-magnon interactions, the influence of photonic effects has received comparatively little attention. Here, we use numerical simulations to disentangle exciton-magnon coupling from the exciton-mediated magnon-photon interactions observed in optical experiments. By simulating realistic scenarios using parameters determined by experiments, we illustrate the strong dependence of these interactions on photonic interference and dispersion effects near excitonic resonances. Such effects shape the optical response to coherent magnons and make it intrinsically nonlinear in the magnon-induced exciton energy shift. Thermal magnons, which have a particularly pronounced impact on excitons, are found to even produce qualitatively different trends in optical signatures. Depending on weak or strong coupling of excitons and photons, the same exciton-magnon interaction can lead to a redshift of optical modes, a nearly vanishing response, or their blueshift. Finally, we demonstrate steps towards optimizing the multiparameter problem of efficient magnon-photon transduction using a machine-learning approach.

DOI: [10.1103/clk-3tfl](https://doi.org/10.1103/clk-3tfl)

I. INTRODUCTION

Light-matter interactions are often strongly enhanced in materials that host narrow and intense excitonic resonances. When these excitons arise in magnetic materials, their large oscillator strength can in principle amplify magneto-optic effects and provide sensitive optical access to spin dynamics. Recent studies on van der Waals (vdW) magnets have shown that this expectation is well founded: layered magnetic crystals such as CrSBr exhibit unusually strong optical responses to changes in their spin configuration [1–3], suggesting that excitons mediate an efficient coupling between magnons and the photon field.

In conventional magneto-optic materials, phenomena such as the magneto-optic Kerr or Faraday effect originate from small magnetization-dependent corrections to a mostly smooth and weakly dispersive dielectric function [4]. In

this regime, photonic thin-film interference often influences only the magnitude of the optical response, but its sign and spectral structure remain determined by magnetization [5]. Excitonic vdW magnets, however, typically operate in a different regime. In such materials, excitons dominate the dielectric function and strongly modify its dispersion. As a consequence, even modest exciton-magnon interactions can induce large and spectrally structured changes that are further shaped by interference in realistic multilayer geometries. Magneto-optic responses therefore not only reflect the underlying spin physics but also the photonic environment.

A prototypical example of this is the layered antiferromagnet CrSBr [6]. In this material, tightly bound excitons [7] with large oscillator strength [3] give rise to isolated, narrow optical resonances in the near-infrared and optical domain [1,8–10]. As discussed in more detail below, excitons and magnons in CrSBr are intimately coupled with a multitude of consequences for its optical properties. The large oscillator strength of CrSBr excitons, together with the ease of integrating vdW materials into microcavities, has recently also enabled studies of magneto-optics in the strong exciton-photon coupling regime. Initial work highlighted the influence of exciton-photon hybridization [3], photonic anisotropy [11,12], and magnon-induced exciton nonlinearities [13]. Very recent studies have even begun to probe magneto-optic properties of exciton-polariton condensates [14,15]. So far, however, most

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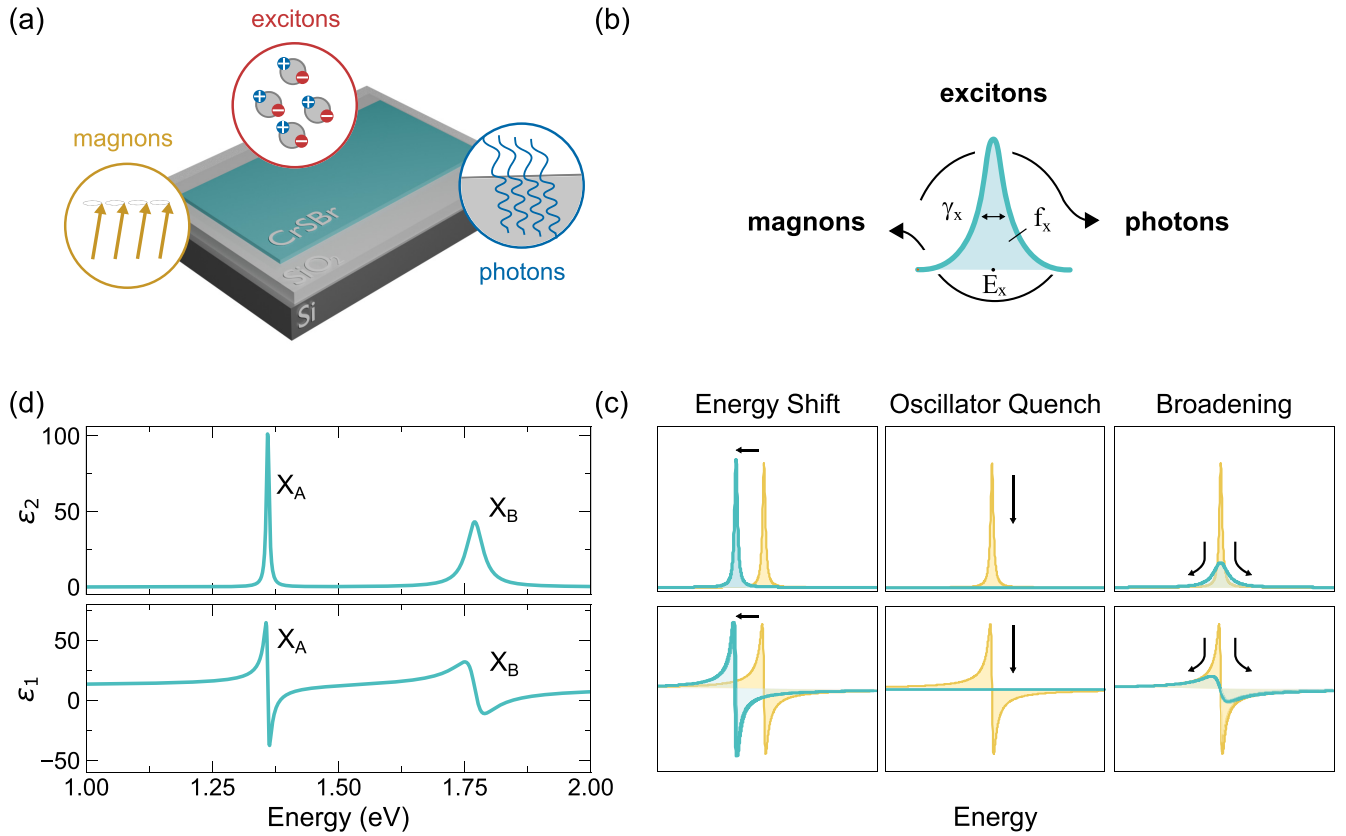


FIG. 1. Role of magnons in the exciton-dominated range of the dielectric function. (a) Sketch of a thin CrSBr crystal on top of a silicon substrate covered by an oxide layer. (b) Magnons couple to photons by changing the exciton resonance. (c) Exciton-magnon interactions can shift, quench, or broaden the exciton resonance. (d) Dielectric function of CrSBr in the near-infrared region modeled using parameters from Ref. [19].

efforts have focused on identifying the microscopic magneto-electric origin of exciton-magnon coupling, leaving the role of photonic interferences [16,17] largely unaddressed.

In this work, we investigate how photonic interference and dispersion influence the magneto-optical response of CrSBr. Using numerical simulations, we reproduce and interpret features observed in recent optical experiments and show that interference in realistic multilayer structures can substantially reshape the apparent signatures of excitons coupled to coherent magnons. We further analyze the impact of thermal magnons on the excitonic dielectric function and demonstrate that their contribution can produce qualitatively different optical outcomes in weakly and strongly coupled exciton-photon regimes. Finally, we illustrate that machine-learning-assisted optimization provides a practical strategy for identifying photonic geometries that maximize the exciton-mediated magnon-photon coupling for transduction.

II. EXCITON-MAGNON INTERACTIONS

To understand the impact of magnetization on optical properties, it can be insightful to consider that magnetic insulators and semiconductors may host a zoo of mutually interacting quasiparticles [Figs. 1(a) and 1(b)]. Magnetic materials studied in the context of magnons are often weakly dispersing in the optical frequency domain [18]. However, when excitons

dominate the optical spectrum, different mechanisms for the coupling of magnons to light become possible.

To illustrate these mechanisms, it is instructive to consider a model dielectric function $\epsilon(E)$ at energies E close to an exciton resonance [Fig. 1(b)], while all other contributions are included in a background relative permittivity ϵ^∞ :

$$\epsilon(E) = \epsilon^\infty - \frac{f_x}{E^2 - E_x^2 + i\gamma_x E}. \quad (1)$$

In this model, magnons can couple to the photon field by shifting the exciton energy E_x , quenching the oscillator strength f_x , or inducing broadening of the exciton linewidth γ_x . The impact of these processes on the dielectric function is schematically depicted in Fig. 1(c).

For light polarized along the b axis, the dielectric function of CrSBr can be approximated by this model. Figure 1(b) shows a plot of its low-temperature dielectric function $\epsilon_b(\omega)$ in the vicinity of both A and B exciton resonances. It is derived from Eq. (1) using parameters determined in recent experiments [19]. Note that surface excitons are omitted in our analysis since their effects are small for crystals with more than a few layers. In the following, we use this dielectric function derived from recent experiments to numerically calculate reflectance spectra of realistic multilayer sample structures via the transfer-matrix method (see Supplemental Sec. S2 [18]). This allows us to analyze the role of photonic interferences in the different optical experiments

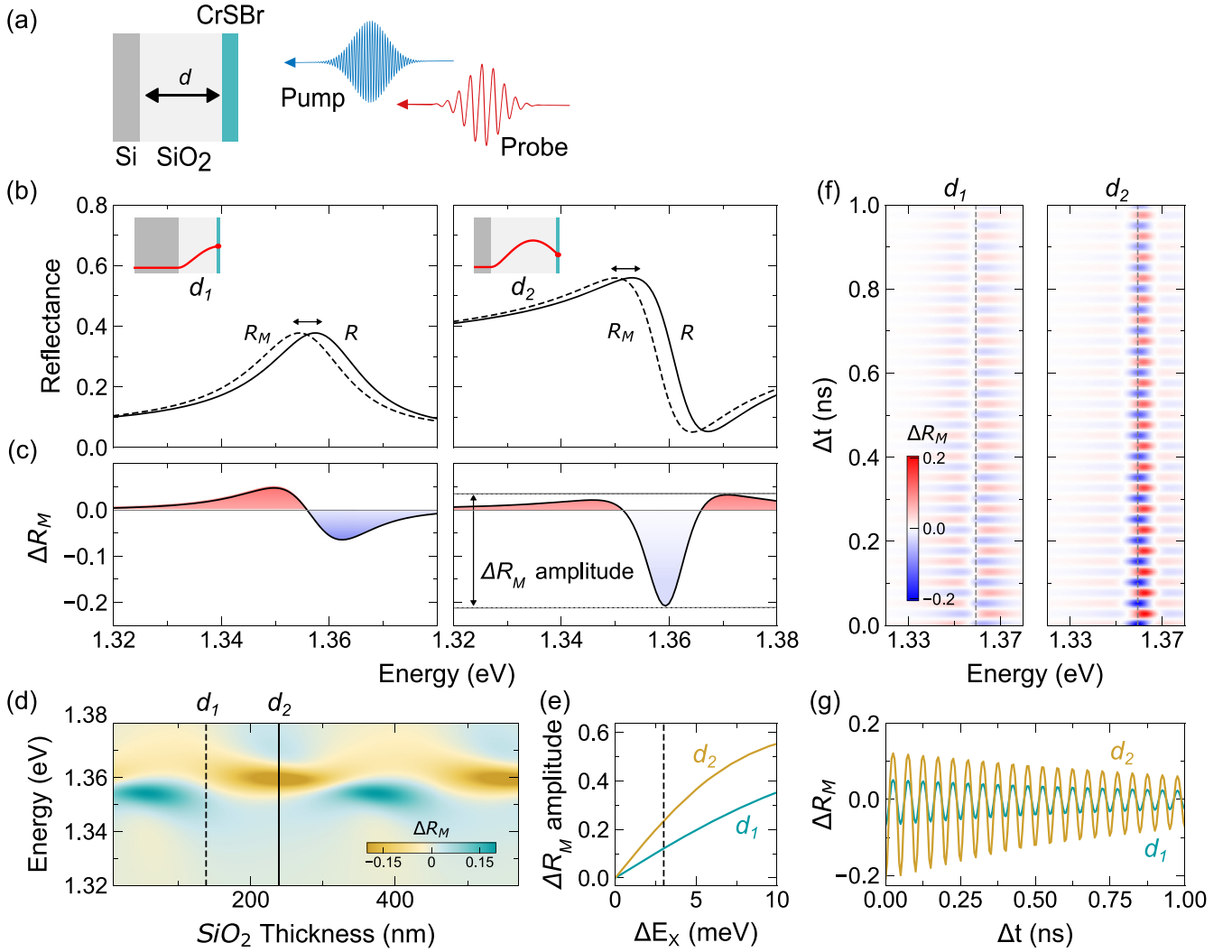


FIG. 2. Coherent magnon-induced reflectance modulation mediated by excitons. (a) Schematic pump-probe measurement of a 5-nm-thick CrSBr crystal on top of a Silicon substrate covered by an oxide of thickness d . (b) Calculated reflectance spectra with (R_M) and without (R) a shift of the exciton energy induced by magnons. Left and right panels compare spectra obtained at oxide thicknesses $d_1 = 140$ nm and $d_2 = 240$ nm. The electric field distribution calculated at $E = E_X$ respectively exhibits a maximum or minimum at the position of the CrSBr crystal for these thicknesses, as indicated in the inset. (c) Magnon-induced reflectance contrast, $\Delta R_M = R_M - R$, and amplitude $|\Delta R_{M,\max} - \Delta R_{M,\min}|$. (d) ΔR_M as a function of energy and SiO₂ thickness. (e) Maximum amplitude of ΔR_M as a function of the magnon-induced exciton energy shift ΔE_X for $d = d_1$ and $d = d_2$. The Dashed line indicates the exciton energy shift used in our simulations. (f) Energy and time dependence of ΔR_M . (g) Line cuts from (f) taken at the maximum amplitude of ΔR_M for $\Delta t = 0$.

demonstrating exciton-magnon coupling. Unless specified otherwise, all spectra below are calculated for a lattice temperature of 20 K, samples with infinite lateral extensions, and light polarized along the crystallographic b axis.

III. COHERENT MAGNONS IN A CANTED SPIN STATE

The most unambiguous optical signature of magnons has been observed in pump-probe measurements [2,3,20,21]. In a seminal experiment, Bae *et al.* showed that ultrafast optical pump pulses can be to create coherent collective oscillations. In a canted state, these oscillations cause a periodic modulation of the interlayer spin angle, which can be detected via changes in the reflectance of time-delayed probe pulses due to changes in the interlayer exciton hopping [2]. This experiment is sketched in Fig. 2(a).

To calculate the magneto-optical response, the coupling of excitons to coherent magnons can be phenomenologically modeled by imposing a damped oscillation of the exciton energy,

$$\Delta E_X(t) = \Delta E_X(0) \cos(\omega_M t) \exp(-t/\tau_M), \quad (2)$$

in the dielectric function of Eq. (1). For our analysis, we use $\Delta E_X(0) = 3$ meV and $\omega_M = 20$ GHz. We also introduce a phenomenological magnon decay characterized by a time constant $\tau_M = 1$ ns, in agreement with experimental observations [2]. Our model thus implicitly assumes both uniform spatial occupation of magnons within the probe volume and the fact that oscillator strength and linewidth of the exciton resonance are not strongly modified by coherent magnons. The latter, in particular, is supported by static reflectance measurements [1,8] indicating that the primary effect of

modifying the interlayer spin angle is a change in the exciton resonance energy E_X . Note that this approach only captures the effects of magnons on the excitonic reflectance response but does not account for additional contributions induced by the absorption of pump pulses. While in a pump-probe experiment, these contributions can lead to background signals, making a quantitative comparison with our simulations difficult, they do not affect the relative effects of photonic interferences on the contribution induced by magnons.

To illustrate the role of photonic interferences in the optical response to coherent magnons, we exemplarily calculate the reflectance of a 5-nm-thick crystal on top of an oxide-covered silicon substrate. Since the thin CrSBr crystal itself does not provide enough optical confinement for self-hybridized polaritons [3], excitons remain weakly coupled to photons. Nonetheless, optical modes are still weakly confined in the oxide layer beneath the CrSBr crystal, as indicated in the insets of Fig. 2(b). These modes are subject to constructive or destructive interference depending on oxide layer thickness. Reflectance in the vicinity of the exciton resonance thus varies strongly with oxide thickness [Fig. 2(d)].

Our discussion focuses on the two cases of constructive ($d_1 = 140$ nm) and destructive ($d_2 = 240$ nm) interference where excitonic reflectance is minimized or maximized, respectively. Figures 2(b) and 2(c) demonstrate the changes in the magnon-induced reflectance contrast, $\Delta R_M = R - R_M$, that occur between them. We find that not only the magnitude, but also the spectral shape of ΔR_M is altered by photonic interferences in the SiO₂ layer. Comparing the two cases, d_1 and d_2 , therefore illustrates the difference between an exciton-mediated magneto-optic responses measured, e.g., in pump-probe experiments, and the underlying exciton-magnon coupling. Figure 2(e) shows another important implication of the fact that the optical response to coherent magnons is mediated by excitons. The response is inherently nonlinear in the magnon-induced exciton shift $\Delta E_X(0)$ due to the nonlinear optical dispersion near exciton resonances. As the spectral shape of the resonances, this nonlinearity depends on oxide thickness. It is, however, only pronounced at large $\Delta E_X(0)$.

Figure 2(f) shows the corresponding time- and energy-resolved pump-probe response of ΔR_M . At destructively interfering optical fields, for $d = d_2$, the amplitude of ΔR_M is more than two times larger. If highly reflective layers such as gold films are part of the stack, this difference even exceeds one order of magnitude (cf. Fig. S2 [18]). In the spectral vicinity of exciton resonances, photonic interferences thus play a significant role for exciton-mediated optical responses to coherent magnons.

IV. THERMAL MAGNONS AND SPIN DISORDER

Magnetic disorder induced by increasing lattice temperature leads to the excitation of incoherent thermal magnons. These magnons randomize the spin alignment not only between adjacent layers but also within individual layers. As a consequence, several parameters of the exciton resonance are modified simultaneously. Experiments indicate that the exciton energy E_X , oscillator strength f_X , and linewidth γ_X all exhibit a pronounced temperature dependence [3,13,19]. The experimentally extracted values [19] used in our simulations

are shown in Figs. 3(a)–3(c). Polynomial fits serve as phenomenological parametrizations.

Several microscopic mechanisms contribute to these trends. Analytical models that incorporate interlayer spin disorder predict a thermal shift of the exciton energy that scales with the density of incoherent magnons in each magnetic sublattice [3]. This mechanism therefore adds a magnetic contribution to the temperature dependence of the exciton resonance beyond the conventional lattice-induced band-gap renormalization. In addition, recent studies report an anomalous temperature dependence of the exciton oscillator strength f_X [11,19,22]. *Ab initio* calculations attribute this effect to reduced hopping amplitudes of electrons and holes along increasingly disordered spin configurations within the layers [19]. Finally, the scattering of excitons by magnons leads to an increase of the exciton linewidth γ_X , consistent with observations from reflectance spectroscopy [19], exciton transport measurements [23], and angle-resolved photoemission experiments [24,25].

Because excitonic resonances dominate the dielectric function of CrSBr in the near-infrared region, these temperature-dependent changes strongly affect the optical response. To illustrate this effect, we calculate the reflectance of a 5-nm-thick CrSBr crystal placed on a Si/SiO₂ substrate for two representative oxide thicknesses corresponding to constructive and destructive interference. The resulting reflectance maps are shown in Figs. 3(d) and 3(e).

The comparison reveals that the experimentally observable reflectance maxima $R_{\max}(T)$ deviate substantially from the intrinsic exciton energy $E_X(T)$. This discrepancy arises because the simultaneous modification of oscillator strength and linewidth alters the interference conditions of the optical field. This is well known yet surprisingly strong in CrSBr due to the large exciton oscillator strength. As highlighted in many previous studies [26–28], it is worth noting that extracting intrinsic excitonic parameters from optical spectroscopy requires quantitative modeling of photonic interference effects.

V. MAGNONS IN THE REGIME OF STRONG EXCITON-PHOTON COUPLING

Recent experiments have demonstrated strong coupling between excitons and photons in CrSBr, resulting in exciton-polariton formation both in intrinsic crystals and in external microcavities [3,22], on photonic lattices [12], and in conventional microcavities [14]. In the following, we investigate how this regime modifies magnon-induced optical responses using transfer-matrix calculations. Our analysis focuses on a prototypical planar microcavity in which a 5-nm-thick CrSBr crystal is embedded in a $\lambda/4$ cavity formed by two planar mirrors [cf. Fig. 4(a)]. The bottom gold mirror is highly reflective (infinite thickness), while the 40-nm-thick top gold mirror is semitransparent. The angle-dependent reflectance spectrum of this microcavity exhibits a lower and an upper polariton branch with varying exciton $|X(\theta)|^2$ and photon $|P(\theta)|^2$ fractions. These fractions can be described using a standard Hopfield model [29] and tuned experimentally via the spacer thickness [cf. Figs. 4(b) and 4(d) inset]. As in most experiments [3,22], we focus on the reflectance response of the lower polariton branch at normal incidence.

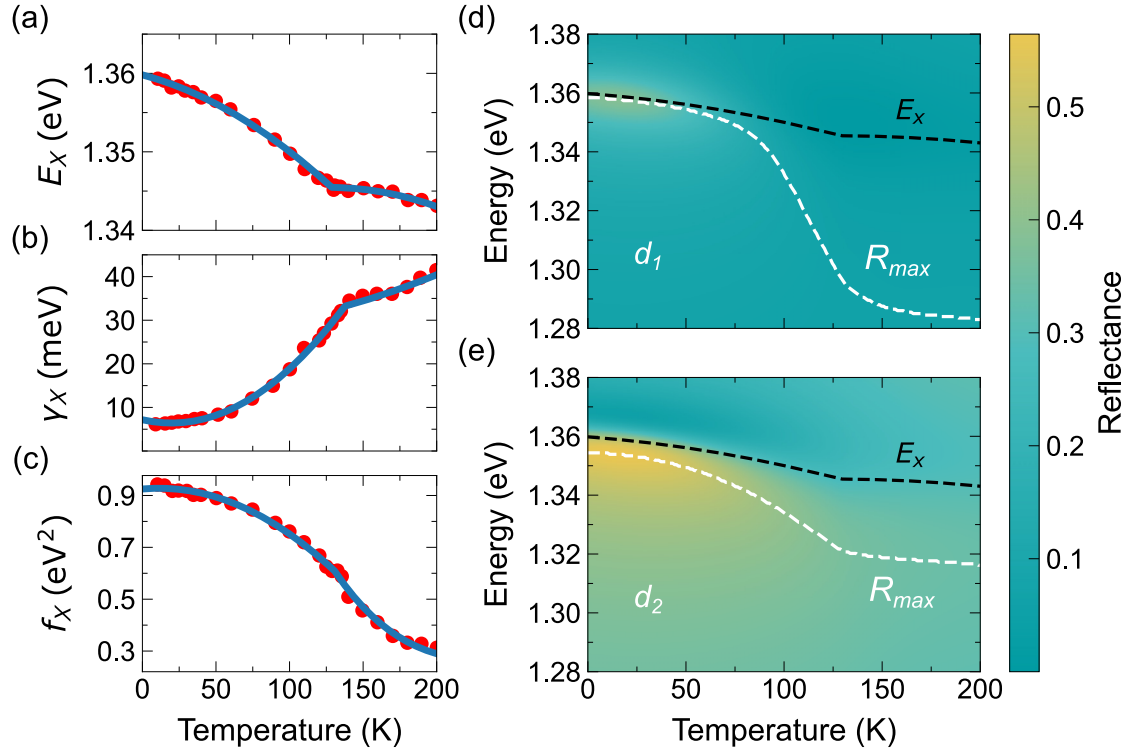


FIG. 3. Temperature dependence of the optical response. (a)–(c) Temperature dependence of excitonic parameters determined by the experiments in Ref. [19] (red dots) approximated by polynomial functions (blue lines) used for calculations. Reflectance of a 5-nm-thick CrSBr crystal placed on a substrate with (d) $d_1 = 140$ nm and (e) $d_2 = 240$ nm oxide thickness. Black dashed lines show $E_X(T)$.

A. Coherent magnons

To model coherent magnons in the strong-coupling regime, we impose the same 3 meV exciton-energy shift as in Sec. III. The calculated reflectance response is shown in Figs. 4(c) and 4(d). Interestingly, the amplitude of the magnon-induced reflectance modulation ΔR_M does not peak for purely excitonic polaritons. Instead, the largest response occurs at an exciton fraction of approximately $|X|^2 \approx 0.8$.

This behavior reflects the hybrid nature of polaritons. The magnon-induced shift originates from the excitonic component, while the photonic component narrows the polariton linewidth and enhances the optical visibility of the resonance shift. As a result, the strongest signal is obtained for polaritons with a mixed exciton-photon character rather than for purely excitonic states.

B. Thermal magnons

The microcavity further enhances the impact of temperature on the optical spectrum of CrSBr. To illustrate this, Fig. 4(e) shows the calculated temperature dependence of the lower-polariton energy for different exciton fractions $|X(0)|^2$. Polaritons with large excitonic content follow the intrinsic redshift of the exciton energy $E_X(T)$ discussed in Sec. IV (cf. Fig. 3). Polaritons with a larger photonic fraction, however, display qualitatively different behavior. For $|X(0)|^2 < 0.7$, the lower-polariton resonance instead shifts towards higher energies with increasing temperature.

This apparently anomalous blueshift arises from two competing effects. Thermal spin disorder reduces the exciton

energy, which by itself would redshift the polariton. At the same time, the oscillator strength decreases with temperature, reducing the Rabi splitting of the exciton-photon system. For lower polaritons, this second effect increases the mode energy and can dominate when the photonic fraction is sufficiently large. Similar behavior has already been reported in several recent experiments [3,14,15,22].

VI. MACHINE-LEARNING-BASED OPTIMIZATION OF MULTILAYER STACKS

Finally, we address the problem of optimizing multilayer stacks towards achieving a maximum amplitude of the magnon-induced reflectance modulation, $|\Delta R_{M,max} - \Delta R_{M,min}|$. This is directly relevant for the magnon-photon transduction schemes proposed in recent works [2,30,31]. In realistic device architectures, the desired transduction would likely proceed through a sequence in which microwave photons excite magnons, magnons interact with excitons, and excitons subsequently couple to near-infrared or optical photons. Efficient signal conversion therefore requires simultaneous optimization of both microwave and optical field distributions. Here, we focus on maximizing the amplitude of ΔR_M .

Already for a simple CrSBr-SiO₂-Si stack, the magnon signal ΔR_M exhibits several local extrema due to its periodic dependence on SiO₂ thickness and the resulting interference effects [cf. Fig. 2(d)]. For a more complex multilayer geometry, identifying the global optimum turns into a high-dimensional optimization problem, a challenge

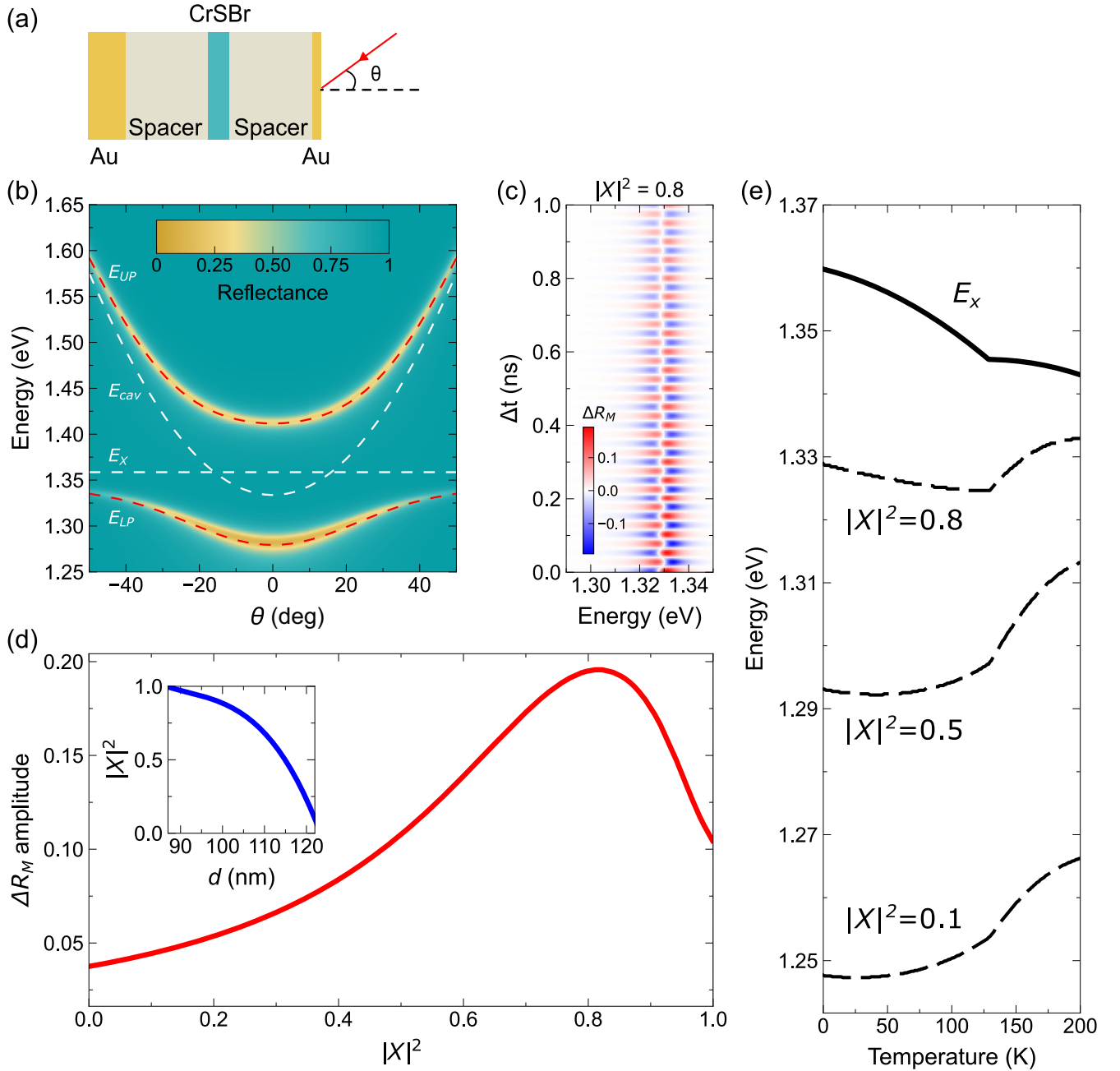


FIG. 4. Magneto-optic responses of a CrSBr microcavity. (a) Microcavity structure. (b) Reflectance calculated for spacer thickness $d = 117$ nm at different angles of incidence, θ . Fits of upper (E_{UP}) and lower (E_{LP}) polariton energies derived from a Hopfield model [29] are plotted as red dashed curves. The Energies of the bare exciton (E_X) and cavity mode (E_{cav}) are indicated by white dashed lines. (c) Transient reflectance induced by coherent magnons for $|X(0)|^2 = 0.8$. (d) Amplitude of ΔR_M as a function of exciton fraction. Inset: Dependence of $|X|^2$ on spacer layer thickness for $\theta = 0^\circ$. (e) Temperature dependence of lower polariton energies for different exciton fractions $|X(0)|^2$.

we tackle by employing the Bayesian optimization structure search (BOSS) algorithm [32]. It efficiently explores the parameter space by iteratively constructing a probabilistic surrogate model of the objective function. New sampling points are then selected based on this model, allowing rapid convergence towards optimal layer configurations. Further details of the optimization procedure, including parameter ranges and constraints, are provided in Supplemental Sec. 2 [18].

Figure 5 shows three prototypical sample structures, calculations of the corresponding electric field profiles for each

of them, and the optimized magnon signal ΔR_M . Case (1) represents a standard sample structure consisting of a 5-nm-thick CrSBr flake on top of a silicon substrate with 285 nm of SiO_2 . It serves as a reference for our optimization approach. The amplitude of ΔR_M in this case reaches a maximum of approximately 0.2. Our optimizer suggests that an enhancement of the magnon signal can be achieved by enclosing the CrSBr layer with a hexagonal boron nitride (hBN) layer on each side [case (2)]. The optimum thicknesses of bottom and top hBN layer are different, as can be seen in Fig. 5(a). In the optimized structure, a maximum amplitude of about 0.3

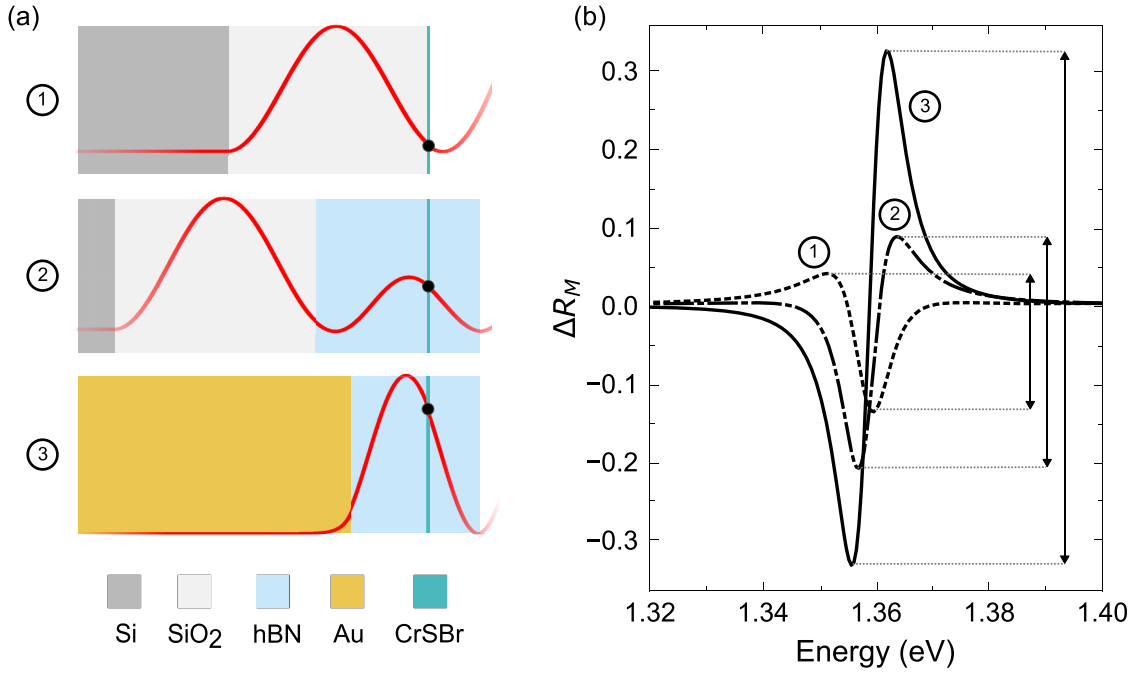


FIG. 5. Optimization of the magnon reflectance response ΔR_M for different multilayer stacks. (a) Schematic of three different prototypical sample structures of experiments. Layer thicknesses in scenarios (2) and (3) are optimized by BOSS. Solid red lines indicate the normalized electric field profile $|\mathcal{E}(z)|^2/|\mathcal{E}|_{\max}^2$ at $E = 1.36$ eV, for each optimized multilayer stack. $|\mathcal{E}|_{\max}$ denotes the field maximum in the calculated region. Black dots mark the optimum position of a 5-nm-thick CrSBr layer for each case. (b) ΔR_M spectra for scenarios (1)–(3). Vertical arrows indicate the amplitude of each ΔR_M curve.

can then be reached. More efficient, however, is the introduction of a gold mirror below the CrSBr layer [case (3)]. By again choosing optimum thicknesses for bottom and top hBN layers, the amplitude is further enhanced, reaching values beyond 0.7.

It is interesting to note that the maximum amplitude of ΔR_M is not achieved by placing CrSBr at the maximum of the electric field. Merely enhancing the interaction of excitons with photonic modes is therefore not the solution our optimizer suggests. Since ΔR_M represents a differential response, its maximum is achieved through a subtle interplay of reflectance resonance amplitude, linewidth, and shift. Additionally, a reduction of the intrinsic exciton linewidth could also significantly enhance the amplitude of ΔR_M . For a linewidth of $\gamma_X \approx 1$ meV, for instance, our optimizer indicates that the theoretical maximum amplitude of $\Delta R_M \approx 2$ comes within reach, enabling most efficient transduction.

VII. CONCLUSION

Excitons in magnetic insulators can mediate a pronounced coupling between magnons and photons, but our analysis shows that the resulting optical signatures depend sensitively on the photonic environment. Unlike in conventional magneto-optics, where the dielectric response is often weakly dispersive, excitonic vdW magnets such as CrSBr operate in a regime where interference, dispersion, and magnetic effects are strongly intertwined. Photonic interference can therefore amplify, suppress, or even invert spectral shifts induced by magnons. Using transfer-matrix simulations, our study finds

that this interplay is relevant in both weak and strong exciton-photon coupling regimes and that thermal magnons and spin fluctuations can produce opposite energy shifts depending on the polariton composition. Careful engineering of the photonic environment, combined with machine-learning-based optimization, can substantially enhance the sensitivity of optical measurements to magnons, which may prove valuable for future schemes aiming to transduce magnetic excitations into optical signals in quantum technologies [33–36].

ACKNOWLEDGMENTS

We thank Prof. Alexey Chernikov and Prof. Geoffrey Diederich for valuable discussions. F.D. and G.B. gratefully acknowledge financial support by the German Research Foundation (Deutsche Forschungsgemeinschaft, DFG) via the Emmy Noether Program (Project-ID 534078167) and the Munich Center for Quantum Science and Technology (MC-QST) under Germany's Excellence Strategy – EXC-2111 – 390814868. A.K. acknowledges financial support from the DFG via Spin + X TRR 173-268565370 (project A13). The work of P.R. and M.S. is funded by the DFG under Germany's Excellence Strategy – EXC 2089/2 – 390776260.

DATA AVAILABILITY

All data necessary to reproduce the results presented in this work are available in the accompanying Zenodo repository [37].

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