Localized surface electromagnetic waves in CrI$_3$-based magnetophotonic structures

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Studies of two-dimensional materials has been booming since the successful exfoliation of the monolayer graphene structure [1]. This pioneering work initiated the search for alternative two-dimensional structures of hexagonal lattices with similar electronic, optical, and transport properties for the potential utility of faster information processing. Lately, the experimental activity has resulted in the discovery of other members of this family, including monolayers of transition metal dichalcogenides [2], phosphorene [3, 4], silicene [5–7], germanene [8] and stanene [9] to name a few. Targeted data-mining efforts have also been devoted to the identification of novel two-dimensional materials [10–12], and in fact some of these theoretical predictions have been verified experimentally [13–15].

Of particular importance for the present investigation is the recent discovery of intrinsic ferromagnetism in Cr$_2$Ge$_2$Te$_6$ and CrI$_3$-based van der Waals structures, where magnetism was studied on monolayers using magneto-optical Kerr effect [15, 16]. Such investigations open avenues for the direct applications of this class of materials, for ultrathin magnetic sensors and spin filters of high efficiency [17–22]. For a long time magnetism was not believed to be present in low dimensional magnetic structures, owing to the Mermin-Wagner theorem [23], which in the isotropic Heisenberg model forbids the formation of collinear magnetic ordering in one- and two-dimensional systems, at any finite temperature. Nevertheless, in CrI$_3$ crystals magnetic ordering is observed experimentally, and is demonstrated to be stabilized by the large out-of-plane magnetic anisotropy that relaxes the Mermin-Wagner constraint. The magnetic anisotropy turns out to substantially suppress transverse spin fluctuations resulting in ferromagnetic ordering being clearly detectable at finite temperatures, even in monolayer systems.

Experimental findings on CrI$_3$-based monolayers, including magnetic exchange coupling and magnetic anisotropy, are supported by the results of first-principles calculations [24–26]. The magnetic properties of this system are governed by the Cr$^{3+}$ ions, which are arranged in a two dimensional hexagonal Bravais lattice, while the non-magnetic I$^-$ ions form an octahedral coordination around Cr. The combination of both relativistic spin-orbit coupling of the heavier ligand I atoms, as well as single-ion anisotropy of Cr atoms, leads to the strong magnetic anisotropy of the structure [24, 25, 27–29]. Unfortunately, much less is known regarding electromagnetic properties of chromium triiodide monolayers. To remedy some of this shortcoming, we explore here a monolayer of chromium triiodide, from the perspective of electromagnetic theory with the focus on plasmon excitations. The appearance and propagation of surface electromagnetic waves at the interface between two media has been extensively studied over last decades [30–32]. Initiated by the description of surface waves at the boundary between the metal and dielectric material, their presence has also been confirmed on specific semiconductor/dielectric interfaces, including a whole family of two-dimensional materials [33–35]. In this paper,
Conductivity tensor can be worked out based on the Kubo formula \[ \sigma \approx \frac{1}{c^2} \int \frac{	ext{d} \omega}{\pi} \chi^{\text{ab}}(\omega) \]

(1) A proper account of trigonal symmetry of a honeycomb lattice suggests only three components to be independent, namely \( \sigma_{0}(\omega) \equiv \sigma_{xx}(\omega) = \sigma_{yy}(\omega) \), \( \sigma_{xy}(\omega) = -\sigma_{yx}(\omega) \), and \( \sigma_{zz}(\omega) \) with the rest being zero. The results of the GGA+U calculations for in-plane components of the conductivity tensor, \( \sigma_{0}(\omega) \) and \( \sigma_{xy}(\omega) \), are shown in Fig. 1. It should be emphasized that the conductivity of a monolayer of CrI\(_3\) does not differ qualitatively from the behavior of bulk, as reported in Ref. [39], and therefore the results presented in Fig. 1 can be generalized to situations involving thicker CrI\(_3\) layers.

A close inspection of Fig. 1 clearly reveals that for energies \( \omega \lesssim 1.2 \text{ eV} \) the transverse component of the conductivity tensor \( \sigma_{xy}(\omega) \) is one-two orders of magnitude smaller than the longitudinal one. In addition to that, it is interesting to note that for small energies, the imaginary part of the diagonal component of the conductivity tensor is negative while the real part is small. These are conditions that might give rise to the surface plasmon wave generation in CrI\(_3\) layer, and this possibility will be discussed below.

**Dispersion of surface electromagnetic waves.** Generation, propagation and detection of plasmon excitations, resulting from the coupling between electromagnetic field and charge density waves of a conducting media, are of central importance of the rapidly advancing area of plasmonics [31]. In this context, chromium triiodide with its unique magneto-optical properties serves as a candidate for hosting surface electromagnetic waves formed in this monolayer structure. To outline the mechanism underlying the emergence and stability of surface plasmon polaritons in a CrI\(_3\) monolayer, we inspect Maxwell’s equations in the geometry shown schematically in Fig. 2. The magnetic CrI\(_3\) layer of thickness \( d \) is positioned at \( z = 0 \) with the \( \hat{z} \) axis pointing out from an insulating medium represented by a glass prism with constant permittivity \( \varepsilon_1 (z < 0) \). Outside the CrI\(_3\) layer is air with permittivity \( \varepsilon_2 = 1 \). Without loss of generality, we suppose the electromagnetic wave to propagate along the \( \hat{z} \) axis with the propagation constant \( q \) and consider an evanescent solution which decays exponentially along the \( \hat{z} \) axis, \( \propto e^{-|q|z} \).

Being a highly conducting material, as shown in Figs. 1a-b, chromium triiodide monolayers lead to the discontinuity of the magnetic field at the boundary \( z = 0 \), \( \mathbf{H}_2 - \mathbf{H}_1 = \sigma(\omega)\mathbf{E}_2^\text{in} \), while leaving the in-plane components of the electric field \( \mathbf{E}_2^\text{in} = (E_x, E_y) \) unchanged, \( \mathbf{E}_2^\text{in} = \mathbf{E}_2^\text{in} \). Thus, under these specified boundary conditions, one may obtain an expression for the dispersion [34-36]:

\[
\left( \frac{i\sigma_0}{\omega\varepsilon_0} + \frac{\varepsilon_1}{\kappa_1} + \frac{\varepsilon_2}{\kappa_2} \right) \left( \frac{i\sigma_0 k_0 - \kappa_1 - \kappa_2}{c_0} \right) = \left( \frac{\sigma_{xy}}{c_0} \right)^2 ,
\]

where \( k_0 = \omega/c \), while \( \varepsilon_0 \) and \( c \) denote the vacuum permittivity and speed of light respectively, and with \( \kappa_{1,2}^2 = q^2 - \varepsilon_{1,2} k_0^2 \) on either side of the monolayer. In...
by a SiO$_2$ bilayer structure is formed by two thin CrI$_3$ layers separated by a SiO$_2$ layer of thickness $a$, with refractive index $n_{SiO_2}$. Light is injected into the structure via a glass prism at the angle $\alpha$.

FIG. 2. The system under consideration: (a) the CrI$_3$ layer of thickness $d$ is placed on top of a glass prism with permittivity $\varepsilon_1$, while the air permittivity is $\varepsilon_2$. The light is incident at the interface between glass and CrI$_3$ at the angle $\alpha$; (b) the bilayer structure is formed by two thin CrI$_3$ layers separated by a SiO$_2$ layer of thickness $a$, with refractive index $n_{SiO_2}$. Light is injected into the structure via a glass prism at the angle $\alpha$.

FIG. 3. The dispersion relation of surface electromagnetic waves $q(\omega)$ obtained as the solution to Eq. (1). It is clearly visible that in the energy window up to 1.2 eV the imaginary part of propagation constant is almost zero $\text{Im} q(\omega) \approx 0$, favoring the generation of surface plasmon polaritons along the CrI$_3$ boundary.

$$\text{Re} q(\omega)$$

$$\text{Im} q(\omega)$$

0 0.2 0.4 0.6 0.8 1.0 1.2 1.4

Energy $\omega$ (eV)

most negligible impact from TE-mode (the absence of TE-mode is attributed to the lack of magnetic response in the system). Remarkably, upon inverting the plot in Fig. 3 we reproduce the well-known square-root behavior at small momenta, $\omega \propto \sqrt{q}$, with proportionality parameter $21.883 \text{GHz} \cdot \text{m}^{-1/2}$. In what follows, we will explore the field distribution upon numerically solving Maxwell’s equations.

**Electromagnetic modeling.** To prove the presence of surface electromagnetic waves bound by a magnetic monolayer, we simulate the propagation of TM-polarized electromagnetic field through a thin film of chromium triiodide in the Kretschmann-Raether (KR) configuration (Fig. 2). This method is known to be quite ubiquitous in generating surface plasmon polaritons. The standard setup consists of a glass prism and a thin film of a lossy material. In our study the latter is the chromium triiodide layer, of thickness $d$, covered by an insulating medium that has a lower refractive index, compared to the glass prism on the opposite side of the chromium triiodide layer. For incident angles greater than the angle of the total internal reflection, the light reaching the boundary between the prism and the thin film material is converted to the evanescent wave at the other side of the boundary, thus ensuring the coupling to the surface electromagnetic waves. The maximum coupling occurs when the wave vector of the incident light matches the value of the surface plasmon propagation constant. In
the reflectance spectrum, the excitation of the surface plasmon polaritons manifests itself as a dip of the reflectance curve, taking place at higher angles compared to the peak representing the effect of total internal reflection (see Fig. 4). The marked dip of the reflectance curve in Fig. 4 stands for the effect of total absorption of the electromagnetic field energy, which is accompanied by the dissipation in the surface plasmon generation.

To study light propagation through the system described in Fig. 2a, we solve numerically a set of Maxwell’s equations with account for electromagnetic boundary conditions at the surface. We consider a TM-polarized field, incident on the prism at the angle \( \alpha \), that is characterized by the following parameters: \( |H_y| = 1 \text{ A/m} \), the frequency is \( f_0 = 242 \text{ THz} \), and input power 1 W. This frequency has been chosen to make the effect as pronounced as possible in the simulations. The refractive indices on both sides of the chromium triiodide monolayer were taken to be: \( n_1=1.5 \) (for glass) and \( n_2=1 \) (for air). For this energy of the field, the components of the monolayer’s conductivity tensor were calculated to be: \( \sigma_0 = (0.307228 - 1.4240759i) \times 10^5 \text{ \Omega}^{-1} \cdot \text{m}^{-1} \), \( \sigma_{xy} = (0.058205 + 0.369802i) \times 10^4 \text{ \Omega}^{-1} \cdot \text{m}^{-1} \) (see Fig. 1).

A close look at the results of numerical simulation of the reflectance spectrum for different CrI\(_3\) film thicknesses shown in Fig. 4 reveal that the resonance angle of surface plasmon polaritons for glass/CrI\(_3\)/air Kretschmann–Raether configuration ranges from \( \alpha = 43.6^\circ \) to 44.1° for \( d \) between 100 and 250 nm whereas the angle of total internal reflection for the system glass/air is \( \alpha = 41.8^\circ \). Comparing the position of the surface plasmon dip, one can see that the thickness of the CrI\(_3\) layer, \( d \), is an important parameter for surface plasmon generation in the here investigated system. The optimal thickness, resulting in almost zero reflectance value (\( \approx 0.005 \text{ a.u.} \)), equals \( d = 100 \text{ nm} \). This demonstrates a very efficient excitation of surface plasmons at these conditions, while for the thinner CrI\(_3\) layers the surface plasmon resonance is less pronounced because there is not enough thickness to absorb light and excite plasmons. Subsequent variation of the CrI\(_3\) layer thickness (\( d > 100 \text{ nm} \)) results in a narrowing and a decrease of the resonance peak amplitude, which is due to optical losses.

In Fig. 5, we present the color contour plot of the \( y \)-component of the magnetic field, \( H_y \), and its distribution in the \( xz \)-plane for a TM-polarized electromagnetic field that propagates across the system with optimal thickness, \( d = 100 \text{ nm} \), at an incident angle \( \alpha = 44.1^\circ \). The profile of the magnetic field component is clearly revealing an electromagnetic field that is localized at the chromium triiodide layer, with an evanescent nature in the insulating medium.

The symmetric bilayer structure. We also investigated the performance of a surface plasmon resonance in a symmetric bilayer structure, where a SiO\(_2\) layer with thickness \( a \) is sandwiched between two identical layers CrI\(_3\) of thickness \( d=100 \text{ nm} \) (shown in Fig. 3b). We find that when light enters the system under the incident angle \( \alpha \) the surface plasmons can be created on the interface between CrI\(_3\) and air, and the position of the resonance angle increases in the range from 42.4° to 43.1° for a variation of spacer thickness (SiO\(_2\) layer) between 450 and 520 nm. The results of the calculations are shown in Fig. 6. Note that for the excitation of surface plasmons at the outer boundary of the proposed system (i.e. at the CrI\(_3\) layer interfaced between the glass prism and the SiO\(_2\) layer), the electromagnetic wave should propagate through the spacer layer in the form of a guided mode that at the same time leads to the generation of an evanescent wave in the neighbouring low-refractive index medium. Therefore, it is clear for the generation of the spacer thickness \( a \) is optimal when it meets the value of any supported wave guide mode in the SiO\(_2\) layer. Meanwhile, when this condition is not fulfilled (for e.g. \( a = 700 \text{ nm} \), see violet dotted line in Fig. 6), the surface plasmon resonance vanishes from the spectrum.

Conclusion. In this paper, we addressed the properties of surface electromagnetic waves bound by a thin ferromagnet layer of chromium triiodide in the Kretschmann–Raether configuration. Using the conductivity tensor obtained within the Kubo formalism from the \textit{ab initio} calculations, we computed the dispersion relation of surface plasmon polaritons. By a direct numerical solution to a set of Maxwell’s equations we showed that in a rather large energy window these waves can be stabilized, and we have estimated the critical angle which corresponds to the absorption level of charge density waves in CrI\(_3\). It turns out that this state inherits the properties of TM-surface plasmon polariton and its features are controlled by the thickness of CrI\(_3\) layer, leading to the decrease in the effect with increasing layer thickness. We also examined the process of surface plasmon generation in the bilayer CrI\(_3\) structures by considering the CrI\(_3\)/SiO\(_2\)/CrI\(_3\)
FIG. 6. Angular response of the system composed by two CrI$_3$ layers of thickness $d = 100$ nm separated by SiO$_2$ layer with thickness $a$ and refractive index $n_{SiO_2} = 1.44$ (Fig. 2a). The light enters the system via the glass prism at the angle $\alpha$. By varying the spacer thickness, the position of the reflectance minimum shifts from 42.4° at $a = 450$ nm (blue solid-circled line) to 43.1° at $a = 520$ nm (orange dashed line). All curves show a characteristic feature of total internal reflection as a local maximum at $\alpha = 41.8°$. When the guided mode condition on the layer thickness is not fulfilled (e.g. when $a = 700$ nm) the surface plasmon resonance vanishes from the spectrum (violet, dotted line).

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