Fundamental limits for nondestructive measurement of a single spin by Faraday rotation

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Faraday rotation, being a dispersive effect, is commonly considered as the method of choice for nondestructive detection of spin states. Nevertheless Faraday rotation is inevitably accompanied by spin flips induced by Raman scattering, which compromises nondestructive detection. Here, we derive an explicit general relation relating the Faraday rotation and the spin-flip Raman scattering cross sections, from which precise criteria for nondestructive detection are established. It is shown that, even in ideal conditions, nondestructive measurement of a single spin can be achieved only in anisotropic media. Monolayers of transitions metal dichalcogenides are shown to be promising candidates for this purpose.

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I. INTRODUCTION

Encoding information into the spin state of a single electron, nucleus, or atom and reading this information nondestructively constitute some of the main challenges of quantum computing and spintronics. These challenges have motivated strong experimental efforts towards electrical and optical detection of single spin states in semiconductors [1]. Optical detection has been demonstrated by polarized photoluminescence or polarization-dependent absorption [2–4], but these methods are destructive. Dispersive methods like nonresonant Kerr or Faraday rotation can be in principle nondestructive and open a way to quantum nondemolition measurements of a single spin state [5]. Nevertheless it is known that even for dispersive measurements, the probe laser may eventually flip the targeted spin, compromising the nondestructive measurement. This limitation is of fundamental nature, since spin Faraday rotation is inevitably linked to spin-flip Raman scattering [6–8]. This issue has been also addressed in details in the context of spin noise spectroscopy, where the signal can be interpreted either as Faraday or as Raman noise [9,10]. To overcome this problem new schemes for quantum nondemolition measurements have been proposed [11–14]. In practice the conditions for nondestructive measurements can be even more challenging to realize due to nonideal experimental conditions, such as light scattering in the sample substrate, low detector quantum efficiency, mixing of spin states, etc. Since the first detections of a single spin by Kerr or Faraday effect [15,16], and thanks to strong experimental efforts and technological progress, nondestructive measurements with these methods are within reach [17,18].

Until now the fundamental limits imposed by spin-flip Raman scattering (SFRS) on such measurements have not been established quantitatively. This work intends to fill this gap through the derivation of an explicit and general relation between the SFRS and the spin Faraday rotation (SFR) cross sections. The notion of cross section is a very general and convenient way to characterize the probability of scattering, capture, or absorption of light or particles when they interact with atoms, defects, or impurities in solids. As shown in Ref. [19] it is also adapted to characterize the Faraday rotation induced by a spin polarization. Instead, in general the Faraday rotation is characterized by the Verdet constant, which is the proportionality factor between the rotation angle on one side and the interaction length times the magnetic field intensity on the other side. This definition is inconvenient for SFR because the rotation angle is proportional to the spin polarization density rather than the magnetic field. A spin polarization density can be created without applied magnetic field by optical pumping or can appear locally because of a spontaneous spin fluctuation. This property is exploited, for example, in spin noise spectroscopy [20–22]. In these situations it is better to introduce the proportionality factor σ_F between the rotation angle θ_F and spin polarization density J_z such that $\theta_F = \sigma_F J_z \ell$, where ℓ is the interaction length [19]. σ_F has the dimension of a surface and can be considered as a cross section for SFR.

As we show below there exists a direct and general relation between the spin-flip Raman scattering cross section σ_R and σ_F. This relation shows that a quantum nondemolition of a single spin by Faraday rotation is not always feasible even in an ideal experiment.

II. GENERAL RELATIONS BETWEEN CROSS SECTIONS

We consider a transparent dielectric material with spins embedded in it. In these conditions the electric induction can be written as [23]

$$\mathbf{D} = \epsilon' \mathbf{E} + i\mathbf{E} \times (G\mathbf{J}). \tag{1}$$

ϵ' is the real part of the dielectric tensor, and $\mathbf{E}$ is the electric field. In general G is a second rank tensor, which becomes a scalar in optically isotropic media such as atomic vapors or cubic semiconductors. $\mathbf{J}$ stands for the spin density. We will consider below dielectric materials with point group symmetry C_{3v} (relevant for crystals with wurtzite structure or for quantum dots with small in-plane asymmetry) or with

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cubic symmetry. With the high-symmetry axis along z, G takes the form

$$G = \begin{pmatrix} G_1 & 0 & 0 \\ 0 & G_1 & 0 \\ 0 & 0 & G_2 \end{pmatrix}. \quad (2)$$

In the following we will only consider a light beam propagating along z and linearly polarized along x. The field amplitude in the dielectric is then given by $\mathbf{E}(\mathbf{r},t) = E_0\hat{x}\exp[i(\mathbf{k}\cdot\mathbf{r}-\omega t)]$ with $\mathbf{k} = k\hat{z}$, and the light intensity in the dielectric medium is $I_0 = \frac{1}{2}\epsilon_0 E_0^2 cn$, where n is the refractive index.

To calculate the Faraday rotation angle θ_F one first considers that $\mathbf{J} = J\hat{z}$ is time independent. The solution of the wave equation is well known and gives

$$\theta_F = \frac{\omega G_2 J\ell}{2cn\epsilon_0}. \quad (3)$$

From the definition of the Faraday rotation cross section [19] one obtains

$$\sigma_F = \frac{\omega G_2}{2cn\epsilon_0}. \quad (4)$$

Let us now calculate the spin-flip Raman scattering (SFRS) cross section σ_R for nonpolarized spins. For this purpose one can consider a small volume v whose dimensions are much smaller than the optical wavelength and that contains N *noninteracting* spins [24]. SFRS occurs because of spin fluctuations. By definition of σ_R the power of light scattered by the N spins in v is given by $P_s = I_0\sigma_R N$. We have thus to calculate P_s. In the volume v the amplitude of the spin-dependent dipole moment induced by the incident field is given by

$$\mathbf{p}(\mathbf{r},t) = i\begin{pmatrix} 0 \\ -G_2E_0J_z(\mathbf{r},t)v \\ G_1E_0J_y(\mathbf{r},t)v \end{pmatrix} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}. \quad (5)$$

The total power emitted by this time-fluctuating dipole is given by

$$P_s = \frac{2}{3}\frac{1}{4\pi\epsilon(c/n)^3}\langle|\ddot{\mathbf{p}}(r,t)|^2\rangle, \quad (6)$$

where $\langle ...\rangle$ denotes time average. Since the spin fluctuations are much slower than the variations of the electromagnetic field one can neglect the time derivatives of $J_{y,z}$. Thus we get

$$P_s = \frac{E_0^2 n\omega^4}{12\pi\epsilon_0 c^3}\left[G_2^2 < (J_z(\mathbf{r},t)v)^2 + G_1^2\langle(J_y(\mathbf{r},t)v)^2\rangle\right]. \quad (7)$$

For N independent and randomly oriented spins we have

$$\langle(J_{y,z}(\mathbf{r},t)v)^2\rangle = \tfrac{1}{3}Ns(s+1), \quad (8)$$

where s is the value of the individual spins. Hence, we obtain the SFRS cross section

$$\sigma_R = \frac{(G_1^2+G_2^2)\omega^4}{18\pi\epsilon_0^2 c^4}s(s+1). \quad (9)$$

By comparing Eqs. (4) and (9) we obtain one of the main results of this paper, which relates σ_R and σ_F

$$\sigma_R = \frac{8\pi}{9}(1+\eta)s(s+1)\left(\frac{n\sigma_F}{\lambda}\right)^2 \quad (10)$$

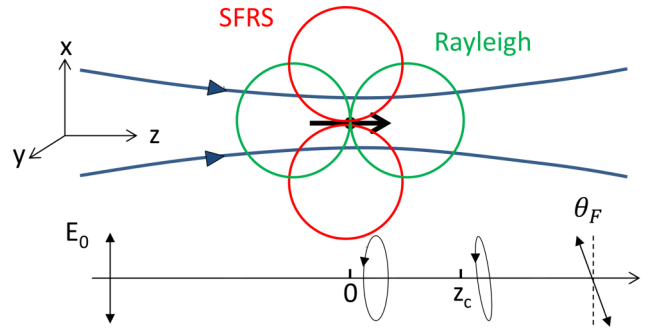

FIG. 1. Up: Schematics of the geometry for dispersive measurement of a single spin by Faraday rotation. The oriented lines represent a focused gaussian beam propagating from left to right, and the arrow represents a spin at the beam waist ($z = 0$), polarized along the z axis. The green (resp. red) circles represent the radiation pattern for Rayleigh scattering (resp. SFRS). In an isotropic medium and for a spin one half the Rayleigh and Raman scattering intensities are exactly equal. Bottom: Illustration of the effect of the Gouy phase shift on the light polarization of the gaussian beam. At $z < 0$ the incident beam is linearly polarized along x. For $\lambda/n \ll z \ll z_c$ the light is elliptically polarized, while for $z \gg z_c$ the light becomes linearly polarized with the polarization rotated by the angle θ_F with respect to the incoming light polarization.

with $\eta = (G_1/G_2)^2$, and $\lambda = 2\pi c/\omega$ is the light wavelength in vacuum.

It may be useful to give the expression of the differential cross section for forward (or backward) scattering, taking into account the radiation pattern of the dipole and that only the y component of the Raman dipole [25] participates to forward scattering. Starting from Eq. (7) one easily gets the differential cross section

$$\left(\frac{d\sigma_R}{d\Omega}\right)_0 = \frac{1}{3}s(s+1)\left(\frac{n\sigma_F}{\lambda}\right)^2, \quad (11)$$

in agreement with Eq. (7) from Ref. [8] for $s = 1/2$.

We consider now a gaussian beam polarized along x and interacting with a single spin $\mathbf{s}$ situated at the position of the beam waist $z = 0$ and in a pure spin-up or spin-down state $s_z = \pm s$ (see Fig. 1). At the beam waist w_0 the intensity of the field decreases with the distance ρ from the beam axis as $I(\rho) = I_0\exp(-(\rho/w_0)^2)$. Since the spin is in a pure state there are no fluctuations along z but only quantum spin fluctuations in the (x, y) plane. Among these, only spin fluctuations in the y direction contribute to SFRS. Hence, in Eq. (7) only the second term of the right-hand side must be kept but with spin fluctuations evaluated for spin-up or spin-down state. Using $s_z = \pm s$ and $\langle s_x^2\rangle = \langle s_y^2\rangle$ one finds $\langle s_y^2\rangle = s/2$. Inserting this value in Eq. (7) one obtains the following expression for the SFRS cross section for a pure spin-up or spin-down state

$$\sigma_{R,\text{pure}} = \frac{4\pi}{3}\eta s\left(\frac{n\sigma_F}{\lambda}\right)^2. \quad (12)$$

Since the Raman dipole is along z there is no forward SFRS in this case (see Fig. 1), in the sense that there is no frequency-shifted scattered light in this direction. However, the static spin component along z induces a dipole parallel to y. The emitted light is not frequency shifted and is cross

polarized with respect to the incident field. It corresponds to spin-Rayleigh scattering and is at the origin of the Faraday effect. The relevant dipole associated with a pure spin state $s_z = \pm s$ is according to Eq. (5) $p_y = \mp iG_2E_0se^{-i\omega t}$. The field emitted along the z axis by this oscillating dipole is given by $E_y(z,t) = \frac{\mu_0}{4\pi r}\ddot{p}_y(t)e^{ikz}$ in the far field ($|z| \gg \lambda/n$). Using Eq. (4) one can express the emitted field as

$$E_y(z,t) = \pm i\sigma_{\rm F}s\frac{n}{\lambda|z|}E_0e^{i(kz-\omega t)}, \tag{13}$$

where $k = n\omega/c$ is the wave vector inside the sample. Besides, the field of the gaussian beam along the z axis is given by

$$E_x(z,t) = \frac{E_0}{\sqrt{1+\left(\frac{z}{z_c}\right)^2}}e^{i(kz-\omega t-\psi(z))}, \tag{14}$$

where $z_c = n\pi w_0^2/\lambda$ is the Rayleigh length, and $\psi(z)$ is the Gouy phase shift. For small rotation the Faraday rotation angle is given by $\mathrm{Re}[E_y(z,t)/E_x(z,t)]$. Close to the spin $\psi(z) \approx 0$, hence the Rayleigh field and the incident field are in phase quadrature. They become in phase only for $z \gg z_c$ where $\psi(z) \to \pi/2$, resulting in a rotation of the polarization plane (see Fig. 1)

$$\theta_{\rm F\pm} = \mp\frac{\sigma_F}{\pi w_0^2}s = \mp\theta_{\rm F}. \tag{15}$$

As expected SFR is proportional to the ratio of $\sigma_{\rm F}$ to the beam cross section. Note however that the rotation angle is not constant across the beam but generally increases off axis. Also SFR is inevitably accompanied by SFRS. Thus there is a finite probability of spin flips induced by light, which limits the possibility of nondestructive measurement.

The total power of elastically scattered light is given by

$$P_{\rm el.} = \frac{G_2^2I_0\omega^4}{6\pi\epsilon_0^2c^4}s^2. \tag{16}$$

As for Raman scattering one gets the corresponding cross section

$$\sigma_{\rm el.} = \frac{8\pi}{3}s^2\left(\frac{n\sigma_F}{\lambda}\right)^2, \tag{17}$$

so that

$$\frac{\sigma_{\rm R,pure}}{\sigma_{\rm el.}} = \frac{\eta}{2s}. \tag{18}$$

III. CONDITION FOR NONDESTRUCTIVE MEASUREMENT OF A SINGLE SPIN

We will consider the case of a spin one half only. For a nondestructive measurement one demands that the acquisition time be short enough to avoid any spin flip induced by inelastic light scattering but long enough to determine the spin state $|s_z\rangle = |\pm\frac{1}{2}\rangle$ from a Faraday effect based measurement. This can be done by measuring the light intensity after a linear polarizer averaged over a large number of photons. But the fundamental detection limit is more rigorously determined by looking at the level of the single spin-single photon interaction, which amounts to detect the single-photon Faraday effect [26,27]. We first consider a statistical measurement based on many photons, and then we will turn to the single-photon detection.

A. Nondestructive measurement with many photons

In the simple approach considered here the induced Faraday rotation is detected with a polarimetric setup consisting of a linear polarizer followed by a detector. The measurement time is such that n_0 photons interact with the spin. As before, we assume that the spin is in a pure state $|\pm\frac{1}{2}\rangle$. The average number of photons scattered due to SFRS is given by

$$n_s = \frac{\sigma_{\rm R,pure}}{\pi w_0^2}n_0. \tag{19}$$

The (perfect) polarizer makes an angle θ with respect to the linear polarization of the in-coming photons. The average number of transmitted photons across the polarizer depends on the spin state and is given by $\langle n_\pm\rangle = n_0\cos^2(\theta \pm \theta_{\rm F})$, where $\theta_{\rm F} = \sigma_{\rm F}/2\pi w_0^2$. The measurement sensitivity is limited by the photon shot noise only, because the intrinsic quantum noise vanishes for pure spin states [28]. In order to determine the spin state, the difference between $\langle n_+\rangle$ and $\langle n_-\rangle$ must be large enough compared to the statistical fluctuations in the detected photon numbers. In such an experiment the spin state can be detected with some fidelity F.

For definiteness we take $\langle n_+\rangle > \langle n_-\rangle$, and we define the fidelity as the probability that the number of detected photons be larger (resp. less) than $\bar{n} = (\langle n_+\rangle + \langle n_-\rangle)/2$ if the spin is in the $|+\frac{1}{2}\rangle$ (resp. $|-\frac{1}{2}\rangle$) state. Thus the fidelity is

$$F(\theta) = \frac{1}{\sqrt{2\pi\langle n_+\rangle}}\int_{\bar{n}}^{+\infty}\exp\left(-\frac{(n-\langle n_+\rangle)^2}{2\langle n_+\rangle}\right)dn, \tag{20}$$

for a Poissonian photon distribution. Taking into account that $\theta_{\rm F} \ll 1$ we find $F(\theta) = \frac{1}{2}(1+\mathrm{erf}(\theta_F\sin(\theta)\sqrt{2n_0}))$. The highest fidelity F is obtained for $\theta \to \pi/2$, i.e., when the polarizer axis is almost perpendicular to the in-coming light polarization.

The minimum number of probe photons necessary to reach a given fidelity is obtained by inverting the above expression, which gives $n_0 = \zeta^2/2\theta_{\rm F}^2$, where $\zeta = \mathrm{erf}^{-1}(2F-1)$. Inserting this value into Eq. (19) and using Eqs. (12) and (15) we obtain

$$n_s > \frac{4\zeta^2\eta}{3}\left(\frac{n\pi w_0}{\lambda}\right)^2. \tag{21}$$

Diffraction imposes $n\pi w_0 \geqslant \lambda$. Hence, the condition that much less than one spin flip should occur during the measurement sets an upper limit to η

$$\eta \ll \frac{3}{4\zeta^2}. \tag{22}$$

Thus the condition depends on the desired fidelity of the measurement. It coincides with the condition given by Eq. (25) for $\zeta = \sqrt{2}$ (see Sec. III B), which corresponds to $F = 0.977$.

B. Nondestructive measurement with a single photon

We now consider the single-photon Faraday effect. The photon-spin interaction leads to a spin-photon entangled state such as

$$|\Psi\rangle = \frac{1}{\sqrt{2}}\left(|-\theta_{\rm F}\rangle\left|+\frac{1}{2}\right\rangle + |\theta_{\rm F}\rangle\left|-\frac{1}{2}\right\rangle\right), \tag{23}$$

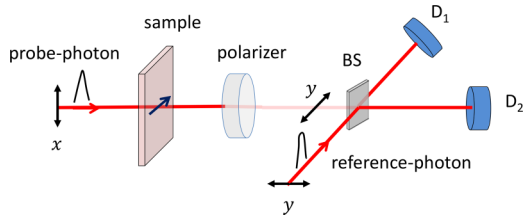

FIG. 2. Principle of a single-shot measurement of a single spin via the Faraday effect using a pair of undistinguishable photons and two-photons detectors D_1 and D_2. One probe photon is sent through the sample and interacts with the spin. The experiment is repeated until a probe photon is transmitted through the polarizer and interferes with a reference photon. A first click on D_1 or D_2 prepares the spin either in the spin-up or spin-down state. Subsequent clicks at the same detector validate the nondestructive measurement.

where $|\pm\theta_F\rangle = \cos(\theta_F)|x\rangle \pm \sin(\theta_F)|y\rangle$ is the photon state. Hence, a phase-sensitive detection of the y component of the photon polarization state projects the spin in a pure state $|\pm\frac{1}{2}\rangle$. If the spin relaxation time is long enough, and if nondestructive measurement is achieved, the outcome of subsequent measurements should always be the same. Figure 2 illustrates a possible phase-sensitive detection setup, using pairs of undistinguishable photons. One photon from each pair (probe photon) is sent though the sample and interacts with the spin, while the other photon (reference photon), which is phase stabilized with respect to the first one is sent on the reference path. After interaction the probe photon state is projected on the y polarization state by the polarizing filter. y-polarized probe photons then interfere with reference photons at the 50:50 beam splitter (BS). A click, corresponding to a two-photons detection [29], will occur either at detector D_1 or at detector D_2 [30]. This prepares the spin in a known pure state. As long as the spin state is conserved, following clicks will always occur at the same detector. At least one click is necessary to detect the spin state. The average number of clicks for n_0 photons incident on the sample is (assuming no loss) $n_{\rm click} = n_0|\langle y|\Psi\rangle|^2 \simeq n_0\theta_F^2$ for small rotation angles. Besides, the number of spin flips due to SFRS in the same time interval is $n_{\rm sf} = n_0\sigma_{\rm R,pol}/\pi w_0^2$. A nondestructive measurement requires both $n_{\rm sf} \ll 1$ and $n_{\rm click} \geqslant 1$. Using Eqs. (15) and (12) we get

$$\frac{8\eta}{3}\left(\frac{n\pi w_0}{\lambda}\right)^2 \ll 1. \tag{24}$$

Taking into account that diffraction imposes $\pi w_0 \geqslant \lambda/n$, we finally obtain the following condition for a nondestructive measurement

$$\eta \ll \tfrac{3}{8}, \tag{25}$$

which can be satisfied only in anisotropic media. This is consistent with the fact that for isotropic media SFRS occurs with the same probability as spin-Rayleigh scattering [see Eq. (18)].

IV. RELATION BETWEEN η AND THE SPIN-FLIP RAMAN TENSOR

In order to calculate η we evaluate the spin-dependent Raman dipole given by Eq. (5) in the limit of a single spin. Hence, in Eq. (5) we substitute $J_i(\mathbf{r},t)v$ by the spin operator $\frac{\hbar}{2}\hat{\sigma}_i$, where the $\hat{\sigma}_i$ are the Pauli matrices. The spin-flip Raman dipole only couples states having the same orbital. Hence we calculate the dipole between two states $|a\rangle$ and $|b\rangle$ having the same orbital and either time-reversed spin states for Raman scattering (calculation of G_1) or the same spin state for spin-Rayleigh scattering (calculation of G_2). As we considered an incident field polarized along x, the Raman dipole $\langle a|p_y(\mathbf{r},t)|b\rangle$ [resp. $\langle a|p_z(\mathbf{r},t)|b\rangle$] is proportional to the element $\alpha_{yx}(a,b)$ [resp. $\alpha_{zx}(a,b)$)] of the SFRS Raman tensor α. From this we deduce

$$G_1 \propto \frac{\alpha_{zx}(a,b)}{i\langle a|\sigma_y|b\rangle} \tag{26}$$

$$G_2 \propto \frac{\alpha_{yx}(a',b')}{-i\langle a'|\sigma_z|b'\rangle}, \tag{27}$$

where σ_y and σ_z are Pauli matrices, and in each case the pair of states must be chosen to have nonzero denominators. The Raman tensor is given by

$$\alpha_{ij}(a,b) \propto \sum_n \langle b|r_i|n\rangle\langle n|r_j|a\rangle/(E_n - E_a - h\nu), \tag{28}$$

where the sum runs over all intermediate states $|n\rangle$ (we assume $E_n - E_a \simeq E_n - E_b$), the r_i are the cartesian components of the position operator, and $h\nu$ is the incident photon energy (for incoming resonance).

For the calculation of G_1 it is convenient to take $|a\rangle = |\uparrow\rangle$ and $|b\rangle = |\downarrow\rangle$, where the spin states are quantized in the z direction, while for the calculation of G_2 one can take $|a'\rangle = |b'\rangle = |\uparrow\rangle$. With this choice of states the anisotropy parameter η takes the form

$$\eta = \left(\frac{G_1}{G_2}\right)^2 = \left(i\frac{\alpha_{zx}(\uparrow,\downarrow)}{\alpha_{yx}(\uparrow,\uparrow)}\right)^2. \tag{29}$$

A. Application to cubic semiconductors

We now estimate η for cubic semiconductors of zinc-blend structure such as GaAs. The relevant localized electron states can be associated with shallow donor levels, or negatively charged QDs. We assume that the optical response is dominated by the lowest optically active transitions between these localized states and the negatively charged or donor-bound excitons.

Using the matrix elements for the Raman active transitions (Fig. 3) we obtain

$$\alpha_{zx}(\uparrow,\downarrow) \propto \frac{-1}{3}\frac{1}{E_\ell - h\nu} \tag{30}$$

$$\alpha_{yx}(\uparrow,\uparrow) \propto \frac{-i}{2}\frac{1}{E_h - h\nu} + \frac{i}{6}\frac{1}{E_\ell - h\nu}, \tag{31}$$

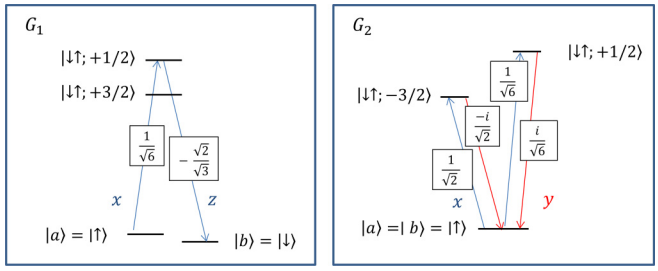

FIG. 3. Left panel: Raman active transitions for spin-flip scattering between two time-reversed spin states deduced from the interband optical matrix elements given in Table I and relevant for the calculation of G_1. The relative amplitudes of the optical matrix elements and the corresponding polarization are given for each active transition. Right panel: Optical transitions relevant to the calculation of G_2.

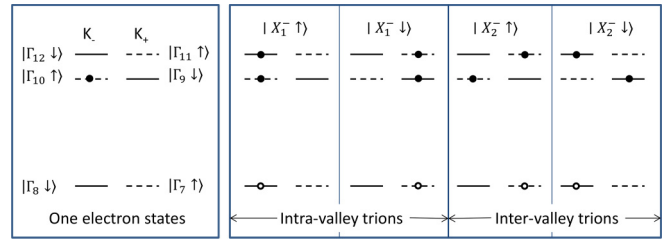

FIG. 4. The left panel shows the one electron states considered in the text, three for each K valley (we adopted the group theory notations of Ref. [37] for the labels of the one electron states). The two lower states are from the top of the valence band; the four upper states are from the bottom of the conduction band. The right panel shows the four allowed trion states involved as intermediate states in the Raman processes. In the labeling of the trion states subscript 1 (resp. 2) refers to intravalley (resp. intervalley) trions.

where E_ℓ and E_h are the energies of the light-hole and heavy-hole transitions, respectively. We deduce

$$\eta = \frac{4}{\left[3\frac{E_\ell - E_h}{E_h - h\nu} + 2\right]^2}. \tag{32}$$

As expected $\eta = 1$ for cubic symmetry (heavy and light-hole states degenerate), and $\eta < 1$ if a uniaxial stress or a confining potential (QDs with cylindrical symmetry) split the heavy-hole and light-hole states. The condition given by Eq. (25) can be fulfilled for large enough splitting between light hole and heavy hole as compared to the detuning from the heavy-hole excitonic resonance, which can be realized in quantum dots.

B. Application to transition metal dichalcogenides (TMDs)

Monolayers of TMDs are direct band gap semiconductors in which spin-valley physics is currently intensively explored [31]. The strong optical anisotropy associated with their 2D structure make them *a priori* good candidates for the nondestructive measurement of the single spin of a resident carrier eventually localized on a shallow donor. Recent DFT calculations show that shallow donors could be obtained by doping in these materials [32,33]. Interestingly, long-lived electron spin dynamics has been demonstrated experimentally in n-type MoS_2 and WS_2 by time-resolved Kerr rotation and ellipticity [34,35].

Figure 4 shows the four lower conduction band states, and the two upper valence band states considered here, as well as the four active negative trion states, which constitute the intermediate states of the Raman processes. We are interested in the detection of a single electron spin in one of the two lower conduction band states ($\Gamma_{10}\uparrow$ or $\Gamma_9\downarrow$), which can be prepared by recombination of a trion. For definiteness let us consider the $\Gamma_{10}\uparrow$ state from the K_- valley.

Table II gives the optical matrix elements for interband transitions. Only spin-conserving transitions are allowed for x-polarized and y-polarized light [36,37], while a non-spin-conserving transition is optically active for the z-polarized light due to spin-orbit mixing [37,38]. The relative amplitude of the non-spin-conserving optical transition is given by β, which represents the fraction of opposite spin admixture in the hole state due to spin-orbit interaction. Due to this mixing Raman spin-flip scattering between $\Gamma_{10}\uparrow$ and $\Gamma_{12}\downarrow$ becomes allowed. From Table II one can easily deduce the active transitions involved in the calculation of G_1 and G_2 (Fig. 5). The calculation of the Raman tensor is straightforward. We find

$$\alpha_{zx}(\uparrow,\downarrow) \propto \frac{\beta}{E_{X_1^-} - h\nu} \tag{33}$$

$$\alpha_{yx}(\uparrow,\uparrow) \propto \frac{-i}{E_{X_1^-} - h\nu} + \frac{i}{E_{X_1^-} + \delta - h\nu}, \tag{34}$$

where $E_{X_1^-}$ is the energy of the optically allowed transition between $X_1^-\uparrow$ and $\Gamma_{10}\uparrow$, and δ is the exchange splitting

TABLE I. Table of optical matrix elements for direct band gap cubic semiconductors. The matrix elements are given for the three components x, y, z of the polarization vector, between the two conduction band spin states (first column) and the four valence band spin states (first raw).

	$\|+3/2\rangle$			$\|+1/2\rangle$			$\|-1/2\rangle$			$\|-3/2\rangle$		
	x	y	z	x	y	z	x	y	z	x	y	z
$\|\uparrow\rangle$	$\frac{1}{\sqrt{2}}$	$\frac{i}{\sqrt{2}}$	0	0	0	$-\frac{\sqrt{2}}{\sqrt{3}}$	$\frac{1}{\sqrt{6}}$	$\frac{-i}{\sqrt{6}}$	0	0	0	0
$\|\downarrow\rangle$	0	0	0	$\frac{1}{\sqrt{6}}$	$\frac{i}{\sqrt{6}}$	0	0	0	$\frac{\sqrt{2}}{\sqrt{3}}$	$\frac{1}{\sqrt{2}}$	$\frac{-i}{\sqrt{2}}$	0

TABLE II. Table of optical matrix elements for TMDs monolayers. The matrix elements are given for the three components x, y, z of the polarization vector, between the four conduction band states (first column) and the two valence band states (first raw).

	$\Gamma_8\downarrow$			$\Gamma_7\uparrow$		
	x	y	z	x	y	z
$\Gamma_{10}\uparrow$	0	0	β	0	0	0
$\Gamma_{12}\downarrow$	1	−i	0	0	0	0
$\Gamma_9\downarrow$	0	0	0	0	0	β
$\Gamma_{11}\uparrow$	0	0	0	1	i	0

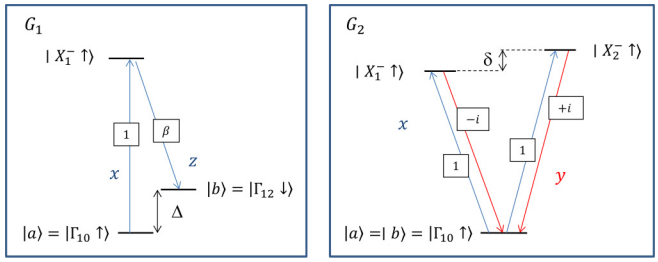

FIG. 5. Left: Raman active spin-flip transition for TMDs illustrated for the K_- valley between two states involved in the calculation of G_1. The relative amplitudes of the optical matrix elements and the corresponding polarization are indicated for each active transition. The intermediate state is an intravalley negative trion. Right: Transitions involved in the calculation of G_2. Both intravalley and intervalley trions split by the exchange interaction contribute to G_2.

between intra- and intervalley trions. Note that the elements of the Raman tensors do not depend on the splitting Δ between $\Gamma_{10}\uparrow$ and $\Gamma_{12}\downarrow$ as long as incoming photon resonance is considered. We thus obtain

$$\eta = \beta^2\left(\frac{E_{X_2^-} - h\nu}{\delta}\right)^2. \tag{35}$$

$\beta \sim 0.03$ in WSe_2 and WS_2 [37,38], and $\delta = 6$ meV in WSe_2 [39]. Hence for a detuning from optical resonance of few meV η can be as small as 0.001. This very small value makes these materials quite promising for nondestructive detection in near-resonant conditions.

V. CONCLUSION

In conclusion we have derived a general relation which connects the SFRS and SFR cross sections, valid in conditions of weak absorption. Using this relation, criteria for nondestructive measurement of a single spin state by Faraday rotation are deduced. These criteria show that nondestructive measurements require a high enough optical anisotropy characterized by the parameter η. The novel 2D materials like TMDs appear to be very promising candidates in this quest. It should be noted that, although a planar cavity amplifies the Faraday rotation, it is useless for nondestructive detection because Raman spin-flip scattering is amplified in such a way that the relation between cross sections remains valid inside the cavity. Finally, the above criteria can be easily adapted for real experiments in order to take into account losses due to light scattering in the substrate and limited quantum efficiency of the detector for example.

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