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Spin Wave Mediated Non-reciprocal Effects in Antiferromagnets

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By using an effective field theory for the electromagnetic interaction of spin waves, we show that, in certain antiferromagnets, the latter induce non-reciprocal effects in the microwave region, which should be observable in the second harmonic generation and produce gyrotropic birefringency. We calculate the various (non-linear) susceptibilities in terms of a few parameters the order of magnitude of which is under control.

1. INTRODUCTION

The response of magnetic materials to electromagnetic fields gives rise to a rich variety of interesting phenomena [1]. In particular, non-reciprocal optical effects in antiferromagnets have received considerable attention during the last years [2, 3]. The possible existence of certain phenomena, like second harmonic generation (SHG) or gyrotropic birefringency (GB), is dictated by the magnetic group of the given material, which may (or may not) allow suitable (non-linear) susceptibilities to be different from zero [4, 5]. The optical wavelengths induce atomic transitions which provide a potential microscopic mechanism to obtain nonvanishing susceptibilities. Indeed this is the case for the observed non-reciprocal effects in Cr_2O_3 [2, 3]. However, alternative mechanisms to produce such effects cannot be ruled out *a priori*, and may even become dominant at certain wavelengths. It is our aim to demonstrate that this is indeed the case for certain antiferromagnets when the electromagnetics fields are in the microwave region. This region is very sensitive to collective magnetic effects which makes a field theoretical description appropriate.

The low temperature low energy properties of antiferromagnets (with spontaneous staggered magnetization) are dominated by spin waves. The spin wave dynamics at low momenta and energy is very much constrained by group theoretical considerations [6]. The symmetry breaking pattern $SU(2) \rightarrow U(1)$ tells us that the spin waves must transform under a non-linear realization of $SU(2)$ [7]. In addition, the space group and time reversal must be respected by the dynamics. The continuum approach ensures that it is enough to consider the rotational part of the space group, namely the point group, and the primitive translations as well as time

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reversal. A systematic description of the spin wave dynamics fully exploiting the above group theoretical constrains has been provided in [8].

In this paper we apply the general framework described in [8] to work out the electromagnetic response of certain aniferromagnets in the microwave region. We have chosen a crystal with no primitive translations mapping points with opposite magnetisations. The point group is taken to be $\overline{3}m$, but repeating the analysis for any other point group is straightforward. This choice is motivated by the Cr_2O_3 crystal, which shows interesting non-reciprocal effects in the optical region as mentioned before. We have calculated the linear and non-linear electric and magnetic susceptibilities, which turn out to depend non-trivially on the frequency of the incoming radiation. In particular non-reciprocal phenomena in the SHG as well as the GB are predicted to occur. These results apply to any antiferromagnet (with spontaneous staggered magnetisation) of arbitrary spin and crystal point group $\overline{3}m$ such that the magnetic ions lie on the z-axis and no primitive translation mapping point with opposite magnetisation exists.

In order to simplify the notation we will take $\hbar = c = 1$, which leads to a relativistic notation. So $x = (t, \mathbf{x})$ and $q = (\omega, \mathbf{k})$. Subindices $\mu = 0, 1, 2, 3$, where the first one represents the time component. Furthermore we work with holomorphic coordinates $z = x + iy$ and $\bar{z} = x - iy$. We distribute the paper as follows. In Section 2 we briefly review some basic aspects of electromagnetic wave propagation in media in order to explain the appearance of non-reciprocal effects in SHG as well in GB. In Section 3 we quickly review the framework described in full detail in [8]. In Section 4 we present the effective lagrangian. In Section 5 we work out the effective action which describes the response to the electromagnetic field and give the (non-linear) electric and magnetic susceptibilities. Section 6 is devoted to a discussion. Finally, in the Appendix we list all the terms which are not displayed in Sections 4 and 5 in order to make the presentation simpler.

2. ELECTROMAGNETIC WAVES PROPAGATION IN MEDIA

It is the aim of this section to briefly review some features of electromagnetic wave propagation in media, which are relevant for the rest of the paper, in particular the phenomena of gyrotropic birefringence (GB) and second harmonic generation (SHG), in connection with non-reciprocal effects, which arise due to time reversal violation in the medium.

Let us recall the Maxwell equations in insulating and chargeless media

$$\begin{gathered}\nabla \mathbf{D} = 0 \\ \nabla \times \mathbf{H} = \partial_0 \mathbf{D} \\ \nabla \times \mathbf{E} = -\partial_0 \mathbf{B} \\ \nabla \mathbf{B} = 0,\end{gathered} \tag{2.1}$$

which are to be supplemented with the constructive equations

$$\begin{aligned} \mathbf{D} &= \mathbf{E} + \mathscr{P} \\ \mathbf{H} &= \mathbf{B} - \mathscr{M}, \end{aligned} \tag{2.2}$$

where $\mathscr{P}$ and $\mathscr{M}$ are the electric and magnetic response of the medium, respectively. $\mathscr{P}$ and $\mathscr{M}$ are functionals of the electric and magnetic fields and, of course, depend on the physical properties of the medium. From the two equations above we obtain

$$\nabla \times \nabla \times \mathbf{E} + \partial_0^2 \mathbf{E} = -(\partial_0^2 \mathscr{P} + \nabla \times \partial_0 \mathscr{M}), \tag{2.3}$$

which is going to be the basic equation in our discussion. Once $\mathscr{P}$ and $\mathscr{M}$ are given this equation describes the propagation of electromagnetic waves in the medium.

For definiteness, consider first the linear response of a homogeneous medium to electric fields only. In this case, the most general form for the electric response is [9]

$$\mathscr{P}^a(z) = \int dx\, \chi^{ab}(z-x)\, E^b(x), \tag{2.4}$$

where x and z are space-time vectors. The tensor $\chi^{ab}(z-x)$ depends not only on time, but also on space coordinates, which characterise the spatial dispersive medium. We shall make the standard assumption that it varies slowly over the medium. This is equivalent to an expansion of the tensor $\chi^{ab}(\omega, \mathbf{k})$ in powers of $\mathbf{k}$ in momentum space. We obtain

$$\mathscr{P}^a(t, \mathbf{z}) = \int dt'\, \chi^{ab}(t-t')\, E^b(t', \mathbf{z}) + \int dt'\, \gamma^{abc}(t-t')\, \partial_c E^b(t', \mathbf{z}) + \cdots. \tag{2.5}$$

The first and second term in the electric response are the polarization and the quadrupolar moment, respectively. Notice that the tensors associated to each order of the multipole expansion depend only on the frequency of the electromagnetic wave.

Let us see next how some terms in the multipole expansion of both the electric and magnetic responses give rize to qualitatively new observable effects. Consider (2.5) together with the leading term in the multipole expansion of the magnetic response (magnetoelectric term)

$$\begin{aligned} \mathscr{P} &= \chi^{ab} E^b + \gamma^{abc}\, \partial_c E^b \\ \mathscr{M} &= \alpha^{ab} E^b. \end{aligned} \tag{2.6}$$

Upon substituting these expressions in (2.3), the quadrupolar and magnetoelectric term give rise to the so-called gyrotropic birefringence as we shall show next.

Consider the plane wave solution, $E^a(x) = E^a(q)\, e^{-iqx} + E^a(-q)\, e^{iqx}$, of Eq. (2.3). Then $E^b(q)$ fulfills

$$[n^2 \delta^{ab} - [\varepsilon^{ab} - n^c(\varepsilon^{acd}\alpha^{db} - i\omega\gamma^{abc})] - n^a n^b]\, E^b(q) = 0, \tag{2.7}$$

where $n^a \equiv k^a/\omega$ gives the propagation direction and its modulus the refraction index (recall that $q = (\omega, \mathbf{k})$). Suppose first that the quadrupolar moment, γ^{abc}, and the magnetoelectric term, α^{bd}, are zero. Non-trivial solutions to this equation arise from the condition that the determinant of the matrix on which the electric field acts vanishes. The anistropy of the permitivity tensor ε^{ab} is generally responsible for this condition to yield two values of the refraction index for each propagation direction, which is known as birefringence. Namely, two different plane waves propagate in each direction with two different polarizations and two different velocities [10]. If the quadrupolar and magnetoelectric terms are restored, they enter Eq. (2.7) through an effective permitivity tensor, $\varepsilon^{ab} - n^c(\varepsilon^{acd}\alpha^{db} - i\omega\gamma^{abc})$, with a linear dependence on the propagation direction, which is known as gyrotropic birefringence [5]. In particular, the equations governing the propagation in directions $\mathbf{n}$ and $-\mathbf{n}$ are different, which implies that the GB is a non-reciprocal effect, since these propagation directions are related by the time reversal.

Let us consider next the non-linear response of the system to electric fields. Thus we have to add to (2.4) new quadratic, cubic, ..., terms [11]

$$\mathscr{P}^a(z) = \int dx\, \chi^{ab}(z-x)\, E^b(x) + \int dx\, dy\, \chi^{abc}(z-x, z-y)\, E^b(x)\, E^c(y) + \cdots . \tag{2.8}$$

In particular, the quadratic term in the above equation leads to the appearance of second harmonic generation. Whenever there exists in the field $E^a(x)$ a contribution of frequency ω the electric response will have in addition two contributions, one of zero frequency and another one of frequency 2ω. Then, the electric response will be, in general, a superposition of plane waves with frequencies the multiple of ω

$$\mathscr{P}^a(z) = \mathscr{P}^a_0(z) + \mathscr{P}^a_q(z) + \mathscr{P}^a_{2q}(z) + \cdots , \tag{2.9}$$

leading, in turn, to a similar superposition for the electric field solution of Eq. (2.3). The contribution to SHG comes from $\mathscr{P}^a_{2q}(z)$ in the expression above, which to lowest order can be written as

$$\begin{aligned} \mathscr{P}^a_{2q}(z) &= P^a(2q)\, e^{-iqz} + P^a(-2q)\, e^{iqz} \\ P^a(2q) &= \chi^{abc}(q, q)\, E^b(q)\, E^c(q). \end{aligned} \tag{2.10}$$

In order to be more explicit consider the non-linear expressions for the electric and magnetic responses,

$$\begin{aligned}\mathscr{P}^a &= \chi^{ab}E^b + \chi^{abc}E^bE^c\\ \mathscr{M}^a &= \mu^{abc}E^bE^c.\end{aligned}\tag{2.11}$$

Once they are introduced in (2.3) we have a complicated non-linear equation. However, the non-linear terms are usually small. Therefore, if we pass the linear part of the response to the l.h.s. we can calculate the electric field solution perturbatively, $\mathbf{E} = \mathbf{E}_{(0)} + \mathbf{E}_{(1)} + \cdots$. $\mathbf{E}_{(0)}$ is the solution of the homogeneous equation (Eq. (2.7) without the quadrupolar and magnetoelectric terms), i.e., a monochromatic plane wave of frequency ω. Then $\mathbf{E}_{(1)}$ follows from the equation

$$\begin{aligned}&\bar{\omega}^2[\bar{n}^2\delta^{ab} - \varepsilon^{ab} + \bar{n}^a\bar{n}^b]\, E^b_{(1)}(\bar{q})\, e^{-i\bar{q}x}\\ &\quad = -(2\omega)^2[\chi^{abc} + n^d e^{ade}\mu^{ebc}]\, E^b_{(0)}(q)\, E^c_{(0)}(q)\, e^{-i2qx}.\end{aligned}\tag{2.12}$$

It is clear that the solution of this equation requires in the l.h.s. an electric field of frequency $\bar{\omega} = 2\omega$. This is called second harmonic generation. Notice, moreover, that the second term in the r.h.s. depends linearly on the direction of the wave number. Then when this term is non-vanishing we have non-reciprocal effects in the second harmonic generation.

In the discussion above we have presented the simplest situations which lead to nonreciprocal effects. In the magnetic materials, as the one we are interested in, $\mathscr{P}$ and $\mathscr{M}$ depend both on the electric and magnetic fields. In this case, since $\mathbf{B} = \mathbf{n} \times \mathbf{E}$, the non-reciprocal effects can be obtained from terms depending on the magnetic field in both the electric and magnetic responses [12, 13]. Furthermore, the (generalized) susceptibilities are constrained by the magnetic point group of the crystal. Since we are considering GB and SHG, which are dynamical effects, only the elements without the time reversal operator of the magnetic group are to be considered [2, 4]. For Cr_2O_3 with the spins aligned in the third direction this is the 32 group. The allowed linear susceptibilities (relevant for the GB) are

$$\begin{aligned}P^z &= \chi_E^{z\bar{z}}E^z + \chi_B^{z\bar{z}}B^z\\ P^3 &= \chi_E^{33}E^3 + \chi_B^{33}B^3\\ M^z &= \gamma_E^{z\bar{z}}E^z + \gamma_B^{z\bar{z}}B^z\\ M^3 &= \gamma_E^{33}E^3 + \gamma_B^{33}B^3,\end{aligned}\tag{2.13}$$

and the bilinear ones (relevant for the SHG)

$$
\begin{aligned}
P^z &= \chi_{EE}^{zzz} E^{\bar z} E^{\bar z} + 2\chi_{EE}^{z\bar z 3} E^z E^3 \\
&\quad + \chi_{EB}^{zzz} E^{\bar z} B^{\bar z} + \chi_{EB}^{z\bar z 3} E^z E^3 + \chi_{EB}^{z3\bar z} E^3 B^z \\
&\quad + \chi_{BB}^{zzz} B^{\bar z} B^{\bar z} + 2\chi_{BB}^{z\bar z 3} B^z B^3 \\
P^3 &= 2\chi_{EE}^{3\bar z z} E^z E^{\bar z} + \chi_{EB}^{3\bar z z}(E^z B^{\bar z} - E^{\bar z} B^z) + 2\chi_{BB}^{3\bar z z} B^z B^{\bar z} \\
M^z &= \gamma_{EE}^{zzz} E^{\bar z} E^{\bar z} + 2\gamma_{EE}^{z\bar z 3} E^z E^3 \\
&\quad + \gamma_{EB}^{zzz} E^{\bar z} B^{\bar z} + \gamma_{EB}^{z\bar z 3} E^z B^3 + \gamma_{EB}^{z3\bar z} E^3 B^z \\
&\quad + \gamma_{BB}^{zzz} B^{\bar z} B^{\bar z} + 2\gamma_{BB}^{z\bar z 3} B^z B^3 \\
M^3 &= 2\gamma_{EE}^{3\bar z z} E^z E^{\bar z} + \gamma_{EB}^{3\bar z z}(E^z B^{\bar z} - E^{\bar z} B^z) + 2\gamma_{BB}^{3\bar z z} B^z B^{\bar z}
\end{aligned}
\tag{2.14}
$$

In the remaining sections we shall calculate the contributions to the generalised susceptibilities above due to the spin wave dynamics. We will start with a local effective lagrangian describing the interaction between spin waves and electromagnetic fields. Upon integrating out the spin waves we obtain a non-local effective action for the electromagnetic fields, which is equivalent to having a free energy [12], taking into account that $L_{int} = -H_{int}$. The electric and magnetic response, and hence all the (generalized) susceptibilities, can be easily obtained as

$$
P^a = \frac{\delta S_{eff}}{\delta E^a}, \qquad M^a = \frac{\delta S_{eff}}{\delta B^a}. \tag{2.15}
$$

3. BUILDING BLOCKS

In this section we present the basic building blocks in the construction of an effective langrangian for the interaction between the spin waves and electromagnetic fields, and their transformations under the relevant symmetries. The method we follow was thoroughly described in a previous article [8]. Here we shall only give a brief overview of it.

As mentioned in the Introduction the spin waves are the lowest lying excited states of the antiferromagnetic ground state associated to the spontaneous symmetry breaking $SU(2) \to U(1)$. This tells us that the associated field, $U(x)$, is an element of the coset space $SU(2)/U(1)$ [7], which transforms under $SU(2)$ as

$$
U(x) \to g U(x)\, h^{\dagger}(g, U), \tag{3.1}
$$

where $g \in SU(2)$ and $h \in U(1)$ is a local ($U(x)$ dependent) element which restores $gU(x)$ to the coset space. If the alignment direction of the local spin is the third direction $U(x)$ can be written as

$$U(x) = \exp\left\{\frac{i\sqrt{2}}{f_\pi}\left[\pi^1(x)\, S^1 + \pi^2(x)\, S^2\right]\right\}, \tag{3.2}$$

where $\pi^i(x)$ are the spin wave fields. These fields in the complex representation have the form $\pi^\pm = (\pi^1 \pm i\pi^2)/\sqrt{2}$ and the generations are written as $S_\pm = S^1 \pm iS^2$.

In addition to the continuous $SU(2)$ transformations the action must be invariant under the space-time transformations. In our case we take the Cr_2O_3 as the underlying crystal in which the spin waves propagate. Cr_2O_3 enjoys the crystallographic point group $\bar{3}m$. The transformation properties of the $U(x)$ field under the $\bar{3}m \otimes T$ elements are

$$\begin{aligned} C_{3z}^+ &: \quad U(x) \to g_3\, U(x)\, h_3^\dagger, & & \\ I &: \quad U(x) \to U(x)\, Ch_I^\dagger, & \qquad C &= e^{-i\pi S^2} \\ \sigma_y &: \quad U(x) \to g_2 U(x)\, h_2^\dagger, & \qquad C^\dagger &= -C. \\ T &: \quad U(x) \to U(x)\, Ch_t^\dagger, & & \end{aligned} \tag{3.3}$$

The non-trivial transformation under I is due to the fact that this particular transformation maps points with opposite local magnetisation in the antiferromagnetic ground state. The primitive translations act trivially on $U(x)$ and have not been displayed.

The spin-orbit is an important interaction which produces a gap in the spectrum of the spin waves because it breaks explicitly the $SU(2)$ symmetry. The breaking part is given by some additional terms in the Heisenberg hamiltonian [14],

$$H = \sum_{<i,j>} J_{ij}\mathbf{S}_i\mathbf{S}_j + \sum_{<i,j>} \mathbf{D}_{ij}(\mathbf{S}_i \times \mathbf{S}_j) + \sum_{<i,j>} M_{ij}^{ab}\, S_i^a\, S_j^b, \tag{3.4}$$

where the tensors D_{ij}^a and M_{ij}^{ab} break the $SU(2)$ symmetry. The order of magnitude of such tensors is $D^a \sim (\Delta g/g)\, J$ and $M^{ab} \sim (\Delta g/g)^2 J$, where for Cr_2O_3, $\Delta g \sim 10^{-2} g$ [15]. In order to introduce them in the effective theory we take their local limit and promote them to sources with proper transformations under $SU(2)$. By combining these sources with the $SU(2)$ generators we obtain objects which transform covariantly under $SU(2)$,

$$\begin{aligned} D_{pq} &\equiv D_{pq}^a S^a \to g D_{pq} g^\dagger \\ M &\equiv M^{ab}(S^a \otimes S^b + S^b \otimes S^a) \to (g \otimes g)\, M(g^\dagger \otimes g^\dagger). \end{aligned} \tag{3.5}$$

Finally they must be fixed to their more general form compatible with the point group symmetry, namely,

$$\begin{aligned} D_{zz} &= D^{-}_{zz} S_{+}, \\ D_{\bar{z}\bar{z}} &= D^{+}_{\bar{z}\bar{z}} S_{-}, \end{aligned} \qquad D^{-}_{zz} = -D^{+}_{\bar{z}\bar{z}}$$

$$\begin{aligned} D_{3z} &= D^{+}_{3z} S_{-}, \\ D_{3\bar{z}} &= D^{-}_{3\bar{z}} S_{+}, \end{aligned} \qquad D^{+}_{3z} = -D^{-}_{3\bar{z}} \tag{3.6}$$

$$M = M^{-+}(S_{+} \otimes S_{-} + S_{-} \otimes S_{+}) + M^{33}(S^{3} \otimes S^{3}).$$

Therefore the objects from which we construct our theory are the spin waves given by $U(x)$, the derivatives, ∂_μ, and the spin-orbit tensors, D^{a}_{ij} and M^{ab}_{ij}. Let us arrange them in a simple form which provides elements with easier transformations properties under $SU(2)$

$$U^{\dagger}(x)\, i\, \partial_\mu U(x) = a^{-}_{\mu}(x)\, S_{+} + a^{+}_{\mu}(x)\, S_{-} + a^{3}_{\mu}(x)\, S^{3}$$

$$U^{\dagger}(x)\, D_{pq} U(x) = d^{-}_{pq}(x)\, S_{+} + d^{+}_{pq}(x)\, S_{-} + d^{3}_{pq}(x)\, S^{3}$$

$$\begin{aligned} (U^{\dagger}(x) \otimes U^{\dagger}(x))\, M(U(x) \otimes U(x)) = {} & m^{--}(x)(S_{+} \otimes S_{+}) \\ & + m^{++}(x)(S_{-} \otimes S_{-}) \\ & + m^{33}(x)(S^{3} \otimes S^{3}) \\ & + m^{-+}(x)(S_{+} \otimes S_{-} + S_{-} \otimes S_{+}) \\ & + m^{-3}(x)(S_{+} \otimes S^{3} + S^{3} \otimes S_{+}) \\ & + m^{+3}(x)(S_{-} \otimes S^{3} + S^{3} \otimes S_{-}). \end{aligned} \tag{3.7}$$

From (3.1) the transformation properties under $SU(2)$ for the coefficients of the generators are

$$\begin{aligned} a^{-}_{\mu}(x) &\to e^{i\theta(x)}\, a^{-}_{\mu}(x) \\ a^{3}_{\mu}(x) &\to a^{3}_{\mu}(x) + \partial_\mu \theta(x) \end{aligned} \tag{3.8a}$$

$$\begin{aligned} & & m^{--}(x) &\to e^{2i\theta(x)} m^{--}(x) \\ d^{-}_{pq}(x) &\to e^{i\theta(x)} d^{-}_{pq}(x), & m^{-3}(x) &\to e^{i\theta(x)} m^{-3}(x) \\ d^{3}_{pq}(x) &\to d^{3}_{pq}(x), & m^{-+}(x) &\to m^{-+}(x) \\ & & m^{33}(x) &\to m^{33}(x), \end{aligned} \tag{3.8b}$$

i.e., the non-linear $SU(2)$ transformation is implemented by a $U(1)_{local}$ transformation. The second transformation in (3.8a) allows us to introduce a covariant derivative $D_\mu \equiv \partial_\mu \pm i a_\mu^3$ acting on $a_\mu^\pm$. Covariant derivatives acting on *ds* or *ms* are redundant and should not be considered (see [8]).

The space-time transformations are given by

$$\xi\colon \{C_{3z}^+, \sigma_y\}\colon \quad \begin{cases} a_\mu^a \to a_{\xi\mu}^a \\ d_{pq}^a \to d_{\xi p \xi q}^a \\ m^{ab} \to m^{ab} \end{cases} \tag{3.9a}$$

$$\xi\colon \{I\}\colon \quad \begin{cases} a_\mu^a \to -a_{\xi\mu}^{\bar{a}} \\ d_{pq}^a \to -d_{pq}^{\bar{a}} \\ m^{ab} \to m^{\bar{a}\bar{b}} \end{cases} \tag{3.9b}$$

$$T\colon \quad \begin{cases} a_\mu^a \to -a_{t\mu}^{\bar{a}} \\ d_{pq}^a \to -d_{pq}^{\bar{a}} \\ m^{ab} \to m^{\bar{a}\bar{b}}, \end{cases} \tag{3.9c}$$

where the symbols $\xi\mu$, ξp, and $t\mu$ represent the transformation of the subindex under the space and time transformations, respectively, together with the corresponding coefficient in each case; the $\bar{a}$ superindex is the conjugate of a.

Next we present the way of introducing the coupling to the electromagnetic field. Since spin waves have no electric charge they couple to the electromagnetic field through the field strength tensor, i.e., direct couplings to the electric and magnetic fields. This kind of coupling does not break the $SU(2)$ symmetry and in order to maintain the space-time symmetry we impose the field **E** transforms like a vector and the field **B** transforms like a pseudovector under the $\bar{3}m$ point group, whereas under time reversal these fields transform as

$$T\colon \begin{cases} E^a \to E^a \\ B^a \to -B^a. \end{cases} \tag{3.10}$$

Since the spin waves are fluctuations of magnetic moments there exists another kind of coupling given by the Pauli term. The Pauli term breaks explicitly the $SU(2)$ symmetry and a source with appropriate transformation properties must be constructed to implement its effect in the effective theory. In the Heisenberg lagrangian with the Pauli interaction, written in the second quantisation language,

$$L = \sum_i \psi^\dagger(x_i)\, i(\partial_0 - i\mu \mathbf{S}\mathbf{B}(x_i))\, \psi(x_i) + \cdots, \tag{3.11}$$

a source $A_0(x) \sim \mu \mathbf{SB}(x)$ can be associated to the Pauli term, which transforms like a connection under time dependent $SU(2)$ transformations,

$$A_0(x) \to g(t)\, A_0(x)\, g^\dagger(t) + ig(t)\, \partial_0 g^\dagger(t), \tag{3.12}$$

such that now the theory will be invariant under time dependent $SU(2)$ transformations. Therefore the effect of the Pauli term is implemented in the effective theory by changing the time derivative by a covariant time derivative,

$$\partial_0 \to D_0 \equiv \partial_0 - iA_0(x), \tag{3.13}$$

and eventually setting $A_0(x) = \mu \mathbf{SB}(x)$. Once this change is performed one has to keep in mind that the $a_0^\pm$ and a_0^3 contain the magnetic field encoded in the covariant time derivative.

At this point the two sources of electromagnetic coupling to spin waves have been considered.

4. EFFECTIVE LAGRANGIAN

Now we are in a position to construct the spin wave interaction with the electromagnetic field for the antiferromagnet. The way we choose to do this is a perturbative one: the derivative expansion. Carrying out this expansion to a given order is meaningful for low energy and momentum with respect to the typical scales of the antiferromagnet, given respectively by the superexchange constant $J \sim 10 meV$ and the inverse of the lattice parameter $1/a \sim 0.1\ \mathring{A}^{-1}$ (the velocity of propagation of the spin waves relates both parameters, $v = Ja$; it has the following value in Cr_2O_3: $v \sim 10^{-4}c$ [15]). The characteristic energy and momentum of the system are given by the external inputs of the electromagnetic fields, which are the same, namely, ω, and therefore the space derivative is highly suppressed with respect to time derivative, $v\partial_i \sim 10^{-4}\partial_0$. The suppression of the spin-orbit tensor has already been given, $D \sim 10^{-2}J$ and $M \sim 10^{-4}J$. Terms proportional to D^2 and M force the local magnetisation to be in the third direction and give rise to an energy gap $\sim 10^{-2}J$ for the spin waves. The amplitude of the electromagnetic field must be constrained for the expansion to make sense. First, we consider the Pauli term. Since it is associated to the time derivative it is suppressed by J. We will assume that the remaining couplings of the electromagnetic field come from vector and scalar potential minimal couplings in a microscopic model. The former is associated to a link and hence suppressed by $1/ea$ whereas the latter is associated to a time derivative and hence suppressed by J/e. Therefore the electric field will be suppressed by J/ea, whereas the magnetic field will be suppressed by $1/ea^2$. The microscopic model may also have non-minimal couplings to the electromagnetic field arising from the integration of higher scales of energy and momentum. These terms would be suppressed by the above mentioned higher scales and will be neglected. In any case, as far as they respect the $SU(2)$ and crystal point group symmetries their only effect is to slightly modify the value of the constants in the effective lagrangian, which are anyway unknown.

Any effect due to spin waves is expected to be enhanced when we approach their energy gap. This is why we shall choose the energy of the electromagnetic wave of that order of magnitude. When in addition the amplitude of the electromagnetic wave is tuned so that $E \sim \partial_0$ the following relative suppressions hold:

$$\begin{aligned} \partial_0,\ eaE,\ d &\sim 10^{-2}J \\ m &\sim 10^{-4}J \\ \mu B &\sim 10^{-5}J \\ v\partial_i,\ eavB &\sim 10^{-6}J. \end{aligned} \tag{4.1}$$

Once the above relations are given we are prepared to construct the relevant effective lagrangian, invariant under $SU(2)$ and space-time transformations given by (3.8), (3.9), and (3.10), for the effect we want to study: Non-reciprocal effects in SHG and GB.

The effective lagrangian at the lowest order in which electromagnetic field appears reads

$$\begin{aligned} S[\pi, E, B] = \int dx f_\pi^2 \{ & a_0^+ a_0^- \\ & + Z_1(d_{zz}^+ d_{\bar{z}\bar{z}}^- + d_{zz}^- d_{\bar{z}\bar{z}}^+) \\ & + Z_2[(d_{3z}^+ d_{zz}^- + d_{3z}^- d_{zz}^+) + (d_{3\bar{z}}^- d_{\bar{z}\bar{z}}^+ + d_{3\bar{z}}^+ d_{\bar{z}\bar{z}}^-) \\ & + Z_3(d_{3z}^+ d_{3\bar{z}}^- + d_{3z}^- d_{3\bar{z}}^+) \\ & + Z_4 d_{zz}^3 d_{\bar{z}\bar{z}}^3 \\ & + Z_5(d_{3z}^3 d_{zz}^3 + d_{3\bar{z}}^3 d_{\bar{z}\bar{z}}^3) \\ & + Z_6 d_{3z}^3 d_{3\bar{z}}^3 \\ & + Z_7 m^{-+} \\ & + Z_8 m^{33} \\ & + Z_9 i[(d_{\bar{z}\bar{z}}^+ a_0^- - d_{\bar{z}\bar{z}}^- a_0^+) E^z - (d_{zz}^- a_0^+ - d_{zz}^+ a_0^-) E^{\bar{z}}] \\ & + Z_{10} i[(d_{3z}^+ a_0^- - d_{3z}^- a_0^+) E^z - (d_{3\bar{z}}^- a_0^+ - d_{3\bar{z}}^+ a_0^-) E^{\bar{z}}] \\ & + Z_{11} E^z E^{\bar{z}} \\ & + Z_{12} E^3 E^3 \}. \end{aligned} \tag{4.2}$$

When we take into acount the Pauli coupling in $a_0^{\pm}$, contributions to SHG arise as 10^{-11} and 10^{-12} effects, and contributions to 10^{-9} appear in the case of GB. As it will be shown these contributions give rise to the desired non-reciprocal effects. Its is important to notice that if a primitive translation mapping points with opposite magnetisation existed the terms with a single time derivative above would not appear in the effective lagrangian.

Our action has been constructed up to third order (10^{-6}) and the contributions to non-reciprocal effects arise only from the Pauli coupling which is much more suppressed in (4.1). Therefore we might expect other contributions at higher orders. This is indeed the case, but in order to keep manageable the number of terms in the main text, these remaining contributions are relegated to the Appendix.

5. ELECTROMAGNETIC FIELD EFFECTIVE INTERACTION

Our purpose is to describe non-reciprocal effects in SHG and GB mediated by spin waves. Spin waves are responsible for an effective interaction of the electromagnetic field giving rise to susceptibility tensors where the properties of the material (spin waves) are encoded.

Hence we realize that spin waves are not to be observed in these experiments and therefore they must be eliminated from our theory. The way to do this is by integrating them out in the functional (path) integral [16] so that the new action depends only on the electromagnetic field. In order to perform the integration we have to write the action explicitly in terms of the spin waves. This is achieved by expanding (3.2), with the result

$$\begin{aligned} S[\pi, E, B] = \int dx[& \partial_0\pi^+\partial_0\pi^- - \Delta^2\pi^+\pi^- \\ & + i\mu(\pi^+\partial_0\pi^- - \pi^-\partial_0\pi^+)\, B^3 \\ & - \tfrac{1}{2}f_\pi[\partial_0\pi^+(\mu B^{\bar{z}} + \lambda E^{\bar{z}}) + \partial_0\pi^-(\mu B^z + \lambda E^z)] \\ & - \tfrac{1}{2}i\mu f_\pi[\pi^+(\mu B^{\bar{z}} + \lambda E^{\bar{z}}) - \pi^-(\mu B^z + \lambda E^z)]\, B^3 \\ & + \tfrac{1}{4}\mu\lambda f_\pi^2(E^z B^{\bar{z}} + E^{\bar{z}} B^z) \\ & + f_\pi^2(b_1 E^z E^{\bar{z}} + b_2 E^3 E^3)], \end{aligned} \tag{5.1}$$

where only the terms contributing to bilinear and trilinear electromagnetic fields in the effective action to be calculated are kept. These terms are the only ones needed to describe the desired effects. The new constants which appear in (5.1) are combinations of those in the previous section. Although we do not know their precise values, their order of magnitude is fixed according to the counting rules given in (4.1). We list them,

$$
\begin{aligned}
f_\pi^2 &\sim \frac{1}{Ja^3}, & \Delta &\sim D \\
b_i &\sim (ea)^2, & \lambda &\sim ea\,\frac{D}{J}.
\end{aligned}
\tag{5.2}
$$

Recal that $D \sim 10^{-2} J$ stands for the size of the spin-orbit term.

Notice that the contributions to non-reciprocal effects to the leading order (10^{-11} and 10^{-12} for SHG and 10^{-9} for GB) come from terms with at most two spin waves, which permits us to perform a gaussian integration in the functional generator. In addition to this, it is worth mentioning that at the order given above the effects are produced at tree level, i.e., without loop contributions.

Once the gaussian integration is carried out a perturbative expansion of the spin waves propagator in the presence of electromagnetic fields has to be made, considering the free spin waves propagator,

$$
P(x-y) = \int \frac{dq}{(2\pi)^4}\, P(\omega)\, e^{-iq(x-y)}, \qquad P(\omega) = \frac{1}{\omega^2 - \Delta^2}, \tag{5.3}
$$

as the unperturbed part, leading to the electromagnetic effective interaction lagrangian

$$
\begin{aligned}
S_{eff}[E, B] = &\int dx\, f_\pi^2 [b_1 E^z E^{\bar z} + b_2 E^3 E^3 + \tfrac{1}{4} \mu\lambda (E^z B^{\bar z} + E^{\bar z} B^z)] \\
&+ \int dx\, dy\, f_\pi^2 [\tfrac{1}{4} \mu\lambda (E^z \partial_0^2 P(x-y)\, B^{\bar z} + B^z \partial_0^2\, P(x-y)\, E^{\bar z})] \\
&+ \int dx\, dy\, f_\pi^2 [-\tfrac{1}{4}\, i\mu\lambda^2 E^z (B^3 \partial_0 P(x-y) + \partial_0 P(x-y)\, B^3)\, E^{\bar z} \\
&- \tfrac{1}{4}\, i\mu^2\lambda [E^z (B^3 \partial_0 P(x-y) + \partial_0 P(x-y)\, B^3)\, B^{\bar z} \\
&+ B^z (B^3 \partial_0 P(x-y) + \partial_0 P(x-y)\, B^3)\, E^{\bar z}]].
\end{aligned}
\tag{5.4}
$$

The arguments of the electromagnetic fields have not been explicitly displayed. They must be understood as the nearest in the closest propagator.

Given the transformations under time reversal (3.10) it is clear that the SHG, terms with three fields, presents non-reciprocal effects due to the interference of different terms. The same is true for the GB since bilinear terms proportional to the magnetic and electric field appear.

From the action above, together with the additional terms given in (A.6), the electromagnetic response of the Cr_2O_3 due to spin waves, leading to non-reciprocal effects in SHG and GB, can be easily obtained using (2.13)–(2.15).

The linear susceptibilities read

$$
\begin{aligned}
\chi_E^{z\bar{z}}(\omega) &= 2b_1 f_\pi^2 \\
\chi_B^{z\bar{z}}(\omega) &= \tfrac{1}{2}\mu\lambda f_\pi^2[1-\omega^2 P(\omega)] \\
\chi_E^{33}(\omega) &= 2b_2 f_\pi^2 \\
\gamma_E^{z\bar{z}}(\omega) &= \tfrac{1}{2}\mu\lambda f_\pi^2[1-\omega^2 P(\omega)].
\end{aligned}
\tag{5.5}
$$

Recall that $\chi_B^{z\bar{z}}(\omega)$ and $\gamma_E^{z\bar{z}}(\omega)$ give rise to the GB. Notice that here this effect is proportional to the gap of the spin wave spectrum, which is in turn due to the spin-orbit interaction.

The susceptibilities contributing to SHG read

$$
\begin{aligned}
\chi_{EE}^{zzz}(\omega,\omega) &= \lambda e_1 f_\pi^2 \omega^3[P(\omega)-4P(2\omega)] + 2\lambda f_1 f_\pi^2 \omega[P(\omega)-P(2\omega)] \\
&\quad + \tfrac{1}{2}\lambda^2 d_1 f_\pi^2 \omega^3 P(\omega)[P(\omega)+2P(2\omega)] \\
\chi_{EE}^{z\bar{z}3}(\omega,\omega) &= \tfrac{1}{2}\lambda e_2 f_\pi^2 \omega^3[P(\omega)+4P(2\omega)] - \lambda e_3 f_\pi^2 \omega^3[P(\omega)-2P(2\omega)] \\
&\quad - \tfrac{1}{2}\lambda f_2 f_\pi^2 \omega[P(\omega)+2P(2\omega)] - \tfrac{3}{2}\lambda^2 d_2 f_\pi^2 \omega^3[P(\omega)\, P(2\omega)] \\
&\quad - 3\lambda^2 c f_\pi^2 \omega^5 P(\omega) P(2\omega) - 3h f_\pi^2 \omega \\
\chi_{EB}^{zzz}(\omega,\omega) &= \lambda g_1 f_\pi^2 \omega[P(\omega)-2P(2\omega)] \\
\chi_{EB}^{z\bar{z}3}(\omega,\omega) &= \tfrac{1}{2}\mu\lambda^2 f_\pi^2 \omega[P(\omega)+2P(2\omega)-6\omega^2 P(\omega)\, P(2\omega)] \\
&\quad - \lambda g_2 f_\pi^2 \omega[P(\omega)+2P(2\omega)] - 6 j_1 f_\pi^2 \omega \\
\chi_{EB}^{z3\bar{z}}(\omega,\omega) &= -2\lambda g_3 f_\pi^2 \omega P(2\omega) + 2 j_2 f_\pi^2 \omega \\
\chi_{BB}^{z\bar{z}3}(\omega,\omega) &= \tfrac{1}{4}\mu^2\lambda f_\pi^2 \omega[P(\omega)+2P(2\omega)-6\omega^2 P(\omega)\, P(2\omega)] \\
\chi_{EE}^{3\bar{z}z}(\omega,\omega) &= -\lambda e_2 f_\pi^2 \omega^3 P(\omega) + \tfrac{1}{2}\lambda e_3 f_\pi^2 \omega^3 P(\omega) \\
\chi_{EB}^{3\bar{z}z}(\omega,\omega) &= -\tfrac{1}{2}\lambda g_3 f_\pi^2 \omega P(\omega) - 2 j_2 f_\pi^2 \omega \\
\gamma_{EE}^{zzz}(\omega,\omega) &= \lambda g_1 f_\pi^2 \omega P(\omega) \\
\gamma_{EE}^{z\bar{z}3}(\omega,\omega) &= -\tfrac{1}{2}\lambda g_3 f_\pi^2 \omega P(\omega) + j_2 f_\pi^2 \omega \\
\gamma_{EB}^{z\bar{z}3}(\omega,\omega) &= \tfrac{1}{2}\mu^2\lambda f_\pi^2 \omega[P(\omega)+2P(2\omega)-6\omega^2 P(\omega)\, P(2\omega)].
\end{aligned}
\tag{5.6}
$$

Notice the non-trivial dependence in ω of the above susceptibilities. (This dependence is slightly more involved if one calculates the general susceptibilities $\chi(\omega,\omega')$ and $\gamma(\omega,\omega')$ since the limit $\omega=\omega'$ produces a few cancellations.) For this to be so it is crucial that no primitive translation mapping points with opposite magnetisation exists. Otherwise only the local terms proportional to j_i would survive. Notice also that the terms proportional to h arise due to the explicit spin-orbit breaking. Even though this term gives rise to a local term in the electromagnetic fields at this order, it also contains explicit interactions with spin waves at higher orders.

Let us finally mention that the terms proportional to k_i in (A.6) give rise to contributions to the quadrupolar momentum [9, 12] which are of the same order as the ones considered above. In fact, the quadrupolar terms give the unique contribution to SHG in a crystal which contains a center of symmetry as it is the case for Cr_2O_3 above the Neel temperature [12]. The associated susceptibilities can be easily calculated. They are local and will not be displayed explicitly.

6. DISCUSSION

We have used an effective field theory for spin waves in an antiferromagnetic material to describe its response to electromagnetic fields in the microwave region. The starting point is a local effective langrangian which fully exploits the fact that spin waves are Goldstone modes of a $SU(2) \rightarrow U(1)$ symmetry breaking pattern together with the crystal space group symmetry and time reversal. By integrating out the spin waves we obtain a non-local effective action which encodes the response of the material to the electromagnetic field. From this effective action the various linear and non-linear electric and magnetic susceptibilities can be immediately obtained. We have given explicitly those relevant to the GB and SHG experiments. These susceptibilities depend on a relatively large number of unknown constants (~ 23) and a microscopic calculation is required to assign definite numbers to them. However, their order of magnitude can be readily established in terms of the typical lattice spacing a and the energy of the first gapped exitation J. Notice also that these susceptibilities present a rather non-trivial dependence on ω, the frequency of the incoming radiation. This dependence cannot be obtained from the magnetic group symmetries alone and it is a direct consequence of the existence of spin waves in an antiferromagnetic crystal where: (i) no primitive translation mapping points with the opposite magnetisations exist, and (ii) spin-orbit effects are sizable.

As mentioned in the Introduction, from group theoretical arguments it has long been known that certain antiferromagnets may support non-reciprocal effects [4, 5]. However, group theoretical considerations alone are unable to indicate any mechanism by which these effects may be realized; they do not even provide an order of magnitude estimate. In Ref. [3] non-reciprocal effects were observed in Cr_2O_3 in the optical region, and in Ref. [2] a theoretical explanation was presented. A micoscopic mechanism leading to such effects was identified in atomic transitions in which spin-orbit interactions and the trigonal field play a crucial role. Here, we have presented a totally different mechanism which also leads to non-reciprocal effects but in the microwave region, namely, the interaction of spin waves with microwave radiation. Since the mechanisms are completely different, the susceptibilities given in [2] hardly have any resemble with ours, exept for the general group theoretical constraints that both of them must fulfil. For instance, the susceptibilities in [2] typically depend on the size of the Cr ions, the energy differences between atomic levels, and matrix elements of perturbations (like spin-orbit terms) between atomic states. Instead, ours typically depend on the lattice spacing, the Heisenberg coupling, and the spin wave energy gap. However, some common features do exist. The spin wave energy gap is due to spin-orbit terms, which are crucial to obtain non-reciprocal effects both for Ref. [2] and for us. In fact, within our approach it is clear that spin-orbit terms are responsible for the spins to point to the third direction, and hence for the magnetic group to be what it is. Also the magnetoelectric susceptibilities in Ref. [2] are proportional to the magnetic moment of the ion and so are ours. Nevertheless, for the optical region it seems that only the susceptibilities γ_E, χ_{EE}, and γ_{EE} are relevant and, then, it is crucial that $\chi_{EE} \sim \gamma_{EE} \neq 0$ for the observation of non-reciprocal effects in the second harmonic generation. In our case all the susceptibilities (i.e., including $\chi_{EB}, \chi_{BB}, \gamma_{EB}$, and γ_{BB}) have the same order of magnitude which provides further observational possibilities. For the sake of comparison, we give below our order of magnitude estimates for the susceptibilities given in [2], assuming the orders of the magnitude that we have been using for the parameters so far. From our γ_E, χ_{EE}, and γ_{EE} we find, adopting the notation of [2], $\alpha^{xx} \sim 10^{-2}$ and $\chi \sim \gamma \sim 10^{-10} CN^{-1}$, which appear to be a few orders of magnitude larger.

We have not been able to locate experimental results in the literature to test our formulas against. We expect them to become available at some point. It would be particularly interesting to be able to browse the microwave region with several frequencies so that the ω dependence in (5.5) and (5.6) could be checked and the free parameters fitted. If the incoming radiation is directed along the third axis then only $\gamma_{EE}^{zzz}(\omega, \omega)$, $\chi_{EB}^{3\bar{z}z}(\omega, \omega)$, $\chi_{EE}^{3\bar{z}z}(\omega, \omega)$, $\chi_{EB}^{zzz}(\omega, \omega)$, and $\chi_{EE}^{zzz}(\omega, \omega)$ are relevant.

Although for definiteness we have focused on the Cr_2O_3 crystal, which has spin 3/2, our results hold for any antiferromagnetic crystal (with spontaneous staggered magnetisation) with crystal point group $\bar{3}m$ and arbitrary spin, as long as no primitive translations mapping points with opposite magnetisation exist. This includes for instance V_2O_3 (spin 1). It is also worth emphasing that the method we have used is general enough to become applicable to any antiferromagnet of any spin and crystal point group, as long as there is spontaneous staggered magnetisation. The allowed terms in the effective lagrangian, however, depend on the particular crystal point group and on the particular distribution of the magnetic ions in the crystal.

APPENDIX A

In this appendix we will present the higher order terms in the effective action (4.2) which contribute to the SHG and GB to the same order as those in (5.1).

Contributions to fourth order (10^{-8});

$$i(a_0^+ D_0 a_0^- - a_0^- D_0 a_0^+)\, E^3 \tag{A.1a}$$

$$\begin{aligned}
&d_{zz}^3(d_{zz}^+ a_0^- + d_{zz}^- a_0^+)\, E^z + d_{\bar z\bar z}^3(d_{\bar z\bar z}^- a_0^+ + d_{\bar z\bar z}^+ a_0^-)\, E^{\bar z}\\
&d_{zz}^3(d_{3\bar z}^+ a_0^- + d_{3\bar z}^- a_0^+)\, E^z + d_{\bar z\bar z}^3(d_{3z}^- a_0^+ + d_{3z}^+ a_0^-)\, E^{\bar z}\\
&d_{3\bar z}^3(d_{3\bar z}^+ a_0^- + d_{3\bar z}^- a_0^+)\, E^z + d_{3z}^3(d_{3z}^- a_0^+ + d_{3z}^+ a_0^-)\, E^{\bar z}\\
&[d_{zz}^3(d_{\bar z\bar z}^+ a_0^- + d_{\bar z\bar z}^- a_0^+) + d_{\bar z\bar z}^3(d_{\bar z\bar z}^- a_0^+ + d_{zz}^+ a_0^-)]\, E^3\\
&[d_{zz}^3(d_{3z}^+ a_0^- + d_{3z}^- a_0^+) + d_{\bar z\bar z}^3(d_{3\bar z}^- a_0^+ + d_{3\bar z}^+ a_0^-)]\, E^3\\
&[d_{3\bar z}^3(d_{3z}^+ a_0^- + d_{3z}^- a_0^+) + d_{3z}^3(d_{3\bar z}^- a_0^+ + d_{3\bar z}^+ a_0^-)]\, E^3;
\end{aligned} \tag{A.1b}$$

to fifth order (10^{-10}),

$$\begin{aligned}
&(d_{zz}^+ D_0 a_0^- + d_{zz}^- D_0 a_0^+)\, E^z E^z + (d_{\bar z\bar z}^- D_0 a_0^+ + d_{\bar z\bar z}^+ D_0 a_0^-)\, E^{\bar z} E^{\bar z}\\
&(d_{3\bar z}^+ D_0 a_0^- + d_{3\bar z}^- D_0 a_0^+)\, E^z E^z + (d_{3z}^- D_0\, a_0^+ + d_{3z}^+ D_0 a_0^-)\, E^{\bar z} E^{\bar z}\\
&(d_{zz}^+ D_0 a_0^- + d_{zz}^- D_0 a_0^+)\, E^{\bar z} E^3 + (d_{\bar z\bar z}^- D_0 a_0^+ + d_{\bar z\bar z}^+ D_0 a_0^-)\, E^z E^3\\
&(d_{3\bar z}^+ D_0 a_0^- + d_{3\bar z}^- D_0 a_0^+)\, E^{\bar z} E^3 + (d_{3z}^- D_0\, a_0^+ + d_{3z}^+ D_0 a_0^-)\, E^z E^3\\
&(d_{zz}^+ a_0^- + d_{zz}^- a_0^+)\, E^{\bar z}\partial_0 E^3 + (d_{\bar z\bar z}^- a_0^+ + d_{\bar z\bar z}^+ a_0^-)\, E^z \partial_0 E^3\\
&(d_{3\bar z}^+ a_0^- + d_{3\bar z}^- a_0^+)\, E^{\bar z}\partial_0 E^3 + (d_{3z}^- a_0^+ + d_{3z}^+ a_0^-)\, E^z \partial_0 E^3
\end{aligned} \tag{A.2a}$$

$$\begin{aligned}
&i(d_{zz}^+ d_{\bar z\bar z}^- - d_{zz}^- d_{\bar z\bar z}^+)(d_{zz}^3 E^z E^z - d_{\bar z\bar z}^3 E^{\bar z} E^{\bar z})\\
&i(d_{3z}^+ d_{3\bar z}^- - d_{3z}^- d_{3\bar z}^+)(d_{zz}^3 E^z E^z - d_{\bar z\bar z}^3 E^{\bar z} E^{\bar z})\\
&i(d_{zz}^+ d_{\bar z\bar z}^- - d_{zz}^- d_{\bar z\bar z}^+)(d_{3\bar z}^3 E^z E^z - d_{3z}^3 E^{\bar z} E^{\bar z})\\
&i(d_{3z}^+ d_{3\bar z}^- - d_{3z}^- d_{3\bar z}^+)(d_{3\bar z}^3 E^z E^z - d_{3z}^3 E^{\bar z} E^{\bar z})\\
&i[(m^{+3} d_{zz}^- - m^{-3} d_{zz}^+)\, E^z E^z - (m^{-3} d_{\bar z\bar z}^+ - m^{+3} d_{\bar z\bar z}^-)\, E^{\bar z} E^{\bar z}]\\
&i[(m^{+3} d_{3\bar z}^- - m^{-3} d_{3\bar z}^+)\, E^z E^z - (m^{-3} d_{3z}^+ - m^{+3} d_{3z}^-)\, E^{\bar z} E^{\bar z}]\\
&i(d_{zz}^+ d_{\bar z\bar z}^- - d_{zz}^- d_{\bar z\bar z}^+)(d_{zz}^3 E^{\bar z} E^3 - d_{\bar z\bar z}^3 E^{\bar z} E^3)\\
&i(d_{3z}^+ d_{3\bar z}^- - d_{3z}^- d_{3\bar z}^+)(d_{zz}^3 E^{\bar z} E^3 - d_{\bar z\bar z}^3 E^{\bar z} E^3)\\
&i(d_{zz}^+ d_{\bar z\bar z}^- - d_{zz}^- d_{\bar z\bar z}^+)(d_{3\bar z}^3 E^{\bar z} E^3 - d_{3z}^3 E^z E^3)\\
&i(d_{3z}^+ d_{3\bar z}^- - d_{3z}^- d_{3\bar z}^+)(d_{3\bar z}^3 E^{\bar z} E^3 - d_{3z}^3 E^z E^3)\\
&i[(m^{+3} d_{zz}^- - m^{-3} d_{zz}^+)\, E^{\bar z} E^3 - (m^{-3} d_{\bar z\bar z}^+ - m^{+3} d_{\bar z\bar z}^-)\, E^z E^3]\\
&i[(m^{+3} d_{3\bar z}^- - m^{-3} d_{3\bar z}^+)\, E^{\bar z} E^3 - (m^{-3} d_{3z}^+ - m^{+3} d_{3z}^-)\, E^z E^3]
\end{aligned} \tag{A.2b}$$

$$
\begin{aligned}
&i(d^3_{zz}E^zB^z - d^3_{\bar{z}\bar{z}}E^{\bar{z}}B^{\bar{z}})\\
&i(d^3_{3\bar{z}}E^zB^z - d^3_{3z}E^{\bar{z}}B^{\bar{z}})\\
&i(d^3_{zz}E^{\bar{z}}B^3 - d^3_{\bar{z}\bar{z}}E^z B^3)\\
&i(d^3_{3\bar{z}}E^{\bar{z}}B^3 - d^3_{3z}E^zB^3)\\
&i(d^3_{zz}E^3B^{\bar{z}} - d^3_{\bar{z}\bar{z}}E^3B^z)\\
&i(d^3_{3\bar{z}}E^3B^{\bar{z}} - d^3_{3z}E^3B^z);
\end{aligned}
\tag{A.2c}
$$

to sixth order (10^{-12}),

$$
\begin{aligned}
&i(d^+_{zz}d^-_{\bar{z}\bar{z}} - d^-_{zz}d^+_{\bar{z}\bar{z}})(E^z\partial_0E^{\bar{z}} - E^{\bar{z}}\partial_0E^z)\,E^3\\
&i[(d^+_{zz}d^-_{3z} - d^-_{zz}d^+_{3z})\,E^z\partial_0E^{\bar{z}} - (d^-_{\bar{z}\bar{z}}d^+_{3\bar{z}} - d^+_{\bar{z}\bar{z}}d^-_{3\bar{z}})\,E^{\bar{z}}\partial_0E^z\,]\,E^3\\
&i(d^+_{3z}d^-_{3\bar{z}} - d^-_{3z}d^+_{3\bar{z}})(E^z\partial_0E^{\bar{z}} - E^{\bar{z}}\partial_0E^z)\,E^3
\end{aligned}
\tag{A.3a}
$$

$$
\begin{aligned}
&(E^z\partial_0E^{\bar{z}} - E^{\bar{z}}\partial_0E^z)\,B^3\\
&E^z\partial_0E^3B^{\bar{z}} - E^3\partial_0E^{\bar{z}}B^z
\end{aligned}
\tag{A.3b}
$$

$$
\begin{aligned}
&(E^z\partial_{\bar{z}}E^z + E^{\bar{z}}\partial_zE^{\bar{z}})\,E^3\\
&E^zE^{\bar{z}}\partial_3E^3\\
&E^zE^{\bar{z}}(\partial_zE^z + \partial_{\bar{z}}E^{\bar{z}})\\
&E^3E^3(\partial_zE^z + \partial_{\bar{z}}E^{\bar{z}}),
\end{aligned}
\tag{A.3c}
$$

where to reduce the number of terms in (A.3b) the homogeneous Maxwell equations, which are satisfied automatically, have been used. It is important to notice that if a primitive translation mapping points with opposite magnetisation existed, the terms in (A.1a), (A.1b), (A.2c), and (A.3a) would not appear.

In spite of the large number of terms, we will see that most of them contribute in the same way to the effective action for the electromagnetic fields.

Indeed, when we expand the terms above in order to make explicit the interaction between the spin waves and the electromagnetic field, the terms below must be added to (5.1) keeping bilinear and trilinear terms in the electromagnetic fields,

$$
\begin{aligned}
\Delta S[\pi, E, B] = \int dx[& ic(\partial_0 \pi^+ \partial_0^2 \pi^- - \partial_0 \pi^- \partial_0^2 \pi^+) E^3 \\
& + id_1(\pi^+ \partial_0 \pi^+ E^z - \pi^- \partial_0 \pi^- E^{\bar{z}}) \\
& + id_2(\pi^+ \partial_0 \pi^- E^3 - \pi^- \partial_0 \pi^+ E^3) \\
& + ie_1 f_\pi(\partial_0 \pi^+ E^z \partial_0 E^z - \partial_0 \pi^- E^{\bar{z}} \partial_0 E^{\bar{z}}) \\
& + ie_2 f_\pi(\partial_0 \pi^+ \partial_0 E^{\bar{z}} E^3 - \partial_0 \pi^- \partial_0 E^z E^3) \\
& + ie_3 f_\pi(\partial_0 \pi^+ E^{\bar{z}} \partial_0 E^3 - \partial_0 \pi^- E^z \partial_0 E^3) \\
& + if_1 f_\pi(\pi^+ E^z E^z - \pi^- E^{\bar{z}} E^{\bar{z}}) \\
& + if_2 f_\pi(\pi^+ E^{\bar{z}} E^3 - \pi^- E^z E^3) \\
& + ig_1 f_\pi(\pi^+ E^z B^z - \pi^- E^{\bar{z}} B^{\bar{z}}) \\
& + ig_2 f_\pi(\pi^+ E^{\bar{z}} B^3 - \pi^- E^z B^3) \\
& + ig_3 f_\pi(\pi^+ E^3 B^{\bar{z}} - \pi^- E^3 B^z) \\
& + ih f_\pi^2(E^z \partial_0 E^{\bar{z}} - E^{\bar{z}} \partial_0 E^z) E^3 \\
& + ij_1 f_\pi^2(E^z \partial_0 E^{\bar{z}} - E^{\bar{z}} \partial_0 E^z) B^3 \\
& + ij_2 f_\pi^2(E^z \partial_0 E^3 B^{\bar{z}} - E^3 \partial_0 E^{\bar{z}} B^z) \\
& + k_1 f_\pi^2(E^z \partial_{\bar{z}} E^z + E^{\bar{z}} \partial_z E^{\bar{z}}) E^3 \\
& + k_2 f_\pi^2 E^z E^{\bar{z}} \partial_3 E^3 \\
& + k_3 f_\pi^2 E^z E^{\bar{z}} (\partial_z E^z + \partial_{\bar{z}} E^{\bar{z}}) \\
& + k_4 f_\pi^2 E^3 E^3 (\partial_z E^z + \partial_{\bar{z}} E^{\bar{z}})].
\end{aligned}
\tag{A.4}
$$

Notice that no terms beyond two spin waves appear in the previous action. This does not change the procedure of gaussian integration carried out in Section 5. The constants in (A.4) have the following order of magnitude:

$$
\begin{aligned}
c &\sim \frac{ea}{J^2}, & d_i &\sim ea \left(\frac{D}{J}\right)^2 \\
e_i &\sim \left(\frac{ea}{J}\right)^2 \frac{D}{J}, & f_i &\sim (ea)^2 \left(\frac{D}{J}\right)^3 \\
g_i &\sim (ea)^2\, Da, & h &\sim (ea)^3\, D^2 \\
j_i &\sim (ea)^3 \frac{a}{J}, & k_i &\sim (ea)^3 \frac{a}{J}.
\end{aligned}
\tag{A.5}
$$

The effective action for the electromagnetic fields in (5.4) must be augmented with the terms

$$
\begin{aligned}
&\Delta S_{eff}[E, B] \\
&= \int dx\, dy\, f_\pi^2 \{ -\tfrac{1}{4} i\lambda e_1 [E^z \partial_0^3 P(x-y) E^z E^z + E^{\bar z} E^{\bar z} \partial_0^3 P(x-y) E^{\bar z}] \\
&\quad - \tfrac{1}{2} i\lambda [E^z \partial_0^2 P(x-y)(e_2 E^{\bar z} \partial_0 E^3 + e_3 \partial_0 E^{\bar z} E^3) \\
&\quad + (e_2 E^z\, \partial_0 E^3 + e_3\, \partial_0 E^z E^3)\, \partial_0^2 P(x-y) E^{\bar z}] \\
&\quad + \tfrac{1}{2} i\lambda [E^z \partial_0 P(x-y)(f_1 E^z E^z + f_2 E^{\bar z} E^3) \\
&\quad + (f_1 E^{\bar z} E^{\bar z} + f_2 E^z E^3)\, \partial_0 P(x-y) E^{\bar z}] \\
&\quad + \tfrac{1}{2} i\lambda [E^z \partial_0 P(x-y)(g_1 E^z B^z + g_2 E^{\bar z} B^3 + g_3 E^3 B^{\bar z}) \\
&\quad + (g_1 E^{\bar z} B^{\bar z} + g_2 E^z B^3 + g_3 E^3 B^z)\, \partial_0 P(x-y) E^{\bar z}]\} \\
&\quad + \int dx\, du\, dy\, f_\pi^2 \{ -\tfrac{1}{4} i\lambda^2 E^z (\partial_0^2 P(x-u)(\mu B^3 + d_2 E^3)\, \partial_0 P(u-y) \\
&\quad + \partial_0 P(x-u)(\mu B^3 + d_2 E^3)\, \partial_0^2 P(u-y))\, E^{\bar z} \\
&\quad - \tfrac{1}{4} i\mu^2 \lambda [E^z (\partial_0^2 P(x-u)\, B^3 \partial_0 P(u-y) + \partial_0 P(x-u)\, B^3 \partial_0^2 P(u-y))\, B^{\bar z} \\
&\quad + B^z (\partial_0^2 P(x-u)\, B^3 \partial_0 P(u-y) + \partial_0 P(x-u)\, B^3 \partial_0^2 P(u-y))\, E^{\bar z}] \\
&\quad + \tfrac{1}{4} i\lambda^2 c E^z (\partial_0^2 P(x-u)\, E^3 \partial_0^3 P(u-y) + \partial_0^3 P(x-u)\, E^3\, \partial_0^2 P(u-y))\, E^{\bar z} \\
&\quad + \tfrac{1}{8} i d_1 \lambda^2 [E^z (\partial_0^2 P(x-u)\, E^z \partial_0 P(u-y) + \partial_0 P(x-u)\, E^z \partial_0^2 P(u-y))\, E^z \\
&\quad - E^{\bar z} (\partial_0^2 P(x-u)\, E^{\bar z} \partial_0 P(u-y) - \partial_0 P(x-u)\, E^{\bar z} \partial_0^2 P(u-y))\, E^{\bar z}]\} \\
&\quad + \int dx\, f_\pi^2 [ih (E^z \partial_0 E^{\bar z} - E^{\bar z} \partial_0 E^z)\, E^3 \\
&\quad + i j_1 (E^z \partial_0 E^{\bar z} - E^{\bar z} \partial_0 E^z)\, B^3 + i j_2 (E^z B^{\bar z} - E^{\bar z} B^z)\, \partial_0 E^3 \\
&\quad + k_1 (E^z \partial_{\bar z} E^z + E^{\bar z} \partial_z E^{\bar z})\, E^3 + k_2 E^z E^{\bar z} \partial_3 E^3 \\
&\quad + k_3 E^z E^{\bar z} (\partial_z E^z + \partial_{\bar z} E^{\bar z}) + k_4 E^3 E^3 (\partial_z E^z + \partial_{\bar z} E^{\bar z})].
\end{aligned}
\tag{A.6}
$$

These contributions to the electromagnetic effective action have been included in our final results for the general susceptibilities in formulas (5.5) and (5.6).

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