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1 **Domain-wall-induced electromagnons in multiferroics**

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Abstract

13

14 Using an atomistic approach, we predict the emergence of new hybrid quasi-particles, namely
15 *domain-wall-induced* electromagnons, that arise from dynamical couplings between magnons and
16 optical phonons in systems possessing ferroelectric domain walls. These quasi-particles induce
17 THz resonances in magnetoelectric responses and preferentially localize either near the domain
18 walls or near the middle of domains. Such behavior is explained through dispersion analysis that
19 allows to track the emergent multi-domain excitations back to single-domain magnetoelectric (ME)
20 modes. The latter, scattered by a periodic array of domain walls, are shown to endow the domain-
21 wall-induced electromagnons with a mixed localized mode and a standing- or propagating-wave
22 characters. Such features can be exploited to reach strikingly large ME conversion and designing
23 more reliable and ultrafast ME devices with less energy consumption and using, e.g., local probes.

24

I. INTRODUCTION

25 Recent observations of magnetoelectric (ME) responses in multiferroics, which charac-
26 terize cross couplings of electric and magnetic properties, has reawakened the interest of
27 scientists and engineers in this territory of research, particularly in the last two decades
28 [1–5]. Such cross couplings open opportunities to, e.g., control electrical polarization by
29 magnetic field or magnetization by electric field. Switching of polarization by magnetic field
30 can be employed technologically in the design of memory devices and magnetic field sensors
31 whereas switching of magnetization by electric field is useful in designing low energy con-
32 sumption storage devices in which data is written electrically and read magnetically [5–11].
33 The existence of excitations other than phonons, such as magnons, in these systems provides
34 more functionalities such as controlling the spin waves by electric field or by optical phonons
35 [6, 12, 13]. The dynamical ME couplings of magnons and phonons, known as electromagnons,
36 have also attracted considerable attention due to their role in magnetocapacitance and mag-
37 netodielectric effects [14–17]. Starting in 2006, experimental reports such as exciting spin
38 waves by light in TbMnO_3 [18] and activating magnetic excitations by applying electric fields
39 in YMn_2O_5 and TbMn_2O_5 [19] kicked off investigations on the origin and mechanism of elec-
40 tromagnons. In particular, it was also experimentally shown that adjusting Terahertz pulses
41 with electromagnons of 0.7 and 1.8 THz in TbMnO_3 can affect its magnetic structure [20].
42 In 2008, electromagnons were proposed to be observed for the first time in single crystals

[21, 22] and thin films [23] of bismuth ferrite BiFeO_3 (BFO). Atomistic methods also predicted electromagnons in antiferromagnetic ferroelectrics [24]. A novel dynamical coupling of magnons with acoustic phonons and optical phonons, coined as electroacoustic magnons, was also suggested in 2019 [25, 26]. Electromagnons can have other benefits. For instance, switching the orientation of domains by applying electric fields on multiferroics is typically of the order of milliseconds [20, 27], although the speed of ferromagnetic or ferroelectric domain reorientation (which is a key in the design of data storage devices) is typically of the order of nanoseconds [20]. In order to improve the operation speed of devices, one can apply high frequency magnetic fields or optical pulses which increase the functionality speed to the order of femtoseconds to picoseconds [28–30] or *coherently excite electromagnons* [31]. Based on the latter possibility, and also to advance fundamental knowledge, it is timely to address a presently unknown issue regarding electromagnons, namely if they can also be preferentially localized within the walls of ferroelectric domains (or near the middle of the domains) rather than “only” equally vibrate in the entire system. If such preferential localizations exist, novel effects can emerge. Examples include new resonances in the magnetoelectric responses and/or a local control of the dynamical conversion between electric and magnetic properties. In order to resolve the aforementioned issues, we performed molecular dynamics (MD) simulations within the GHz-THz regime of applied magnetic fields in BFO materials possessing ferroelectric domain walls. Fourier analysis of temporal evolution of dipole and magnetic moments along with the calculations of quadratic magnetoelectric response of BFO reveals that the existence of domain walls does result in the emergence of *domain-wall-induced electromagnons*, which in turn induce large enhancement of ME response around their resonances. These novel quasi-particles were further found to either be preferentially localized near the domain walls or near the center of the domains (depending on their frequency and direction of propagation of their optical phonon and magnon), as a result of scattering by the periodic array made by the domain walls. Such discoveries have the potential to pave the way for designing original devices. For instance, although the capabilities of magnons as the carriers of information in the field of magnonics is quite promising [32], one of the main challenges is to find a way to control the two-dimensional propagation of the magnons in an energy-efficient fashion, especially in small length scales. A previously proposed approach, which was also realized in experiment recently [33], is to guide the spin-waves in a reconfigurable domain wall as magnonic waveguides. Here, the

75 present work revealing the existence of domain-wall-induced electromagnons strongly sug-
 76 gests that magnonic circuitry can be utilized by activating, *via electric fields*, the magnons
 77 associated with such preferentially localized electromagnons. This should allow to design
 78 novel configurable electronic circuits with much less energy consumption.

79 The article is organized as follows. Section II details the method used here, while results
 80 are presented and discussed in Section III. Finally, Section IV provides a summary of this
 81 work.

82 II. METHODS

83 An effective Hamiltonian framework developed for bismuth ferrite and detailed in Ref.
 84 [34] is employed to perform molecular dynamics simulations for the investigation of the tem-
 85 poral evolution of polarization and magnetization as well as the quadratic magnetoelectric
 86 response of BFO (which is one of the few materials with magnetoelectric functionalities at
 87 room temperature, making it a prime candidate for designing novel devices [10, 35–37]) un-
 88 der the application of magnetic fields with various frequencies from 10 to 1300 GHz. Note
 89 that the applied time-dependent magnetic field can generate an inhomogeneous electric field
 90 which we neglected here in order to merely focus on the ME response rather than the polar-
 91 ization caused by the dielectric response of BFO. In addition, modeling such an electric field
 92 was not feasible in the simulations because of using periodic boundary conditions in all three
 93 Cartesian directions. Note also that our molecular dynamics simulations are performed at
 94 the temperature of 10 K to reduce numerical noises. However, we numerically found that
 95 conducting them at room temperature leads to similar results albeit with lower numerical
 96 resolutions for the magnetoelectric coefficients [25].

97 This presently used effective Hamiltonian scheme encompasses three main energy contri-
 98 butions given by Equation 1 [13, 34, 38]:

$$E^{tot} = E^{FE}(\{\mathbf{u}\}, \{\eta_l\}) + E^{AFD}(\{\mathbf{u}\}, \{\eta_l\}, \{\boldsymbol{\omega}\}) + E^{MAG}(\{\mathbf{m}\}, \{\mathbf{u}\}, \{\eta_l\}, \{\boldsymbol{\omega}\}). \quad (1)$$

99 In Equation 1, local modes $\{\mathbf{u}\}$ are directly related to local electric dipoles. Strain tensor
 100 $\{\eta_l\}$ includes both homogeneous $\{\eta_H\}$ and inhomogeneous $\{\eta_I\}$ strains. The tiltings of
 101 oxygen octahedra (also known as antiferrodistortive or AFD motions) are denoted by $\{\boldsymbol{\omega}\}$,
 102 and the magnetic moments associated with iron ions are denoted by $\{\mathbf{m}\}$. Also, in Equation

103 1, the first term includes the energy of local modes and elastic deformations that can be
 104 written as [39, 40]

$$\begin{aligned}
 E^{FE} = & E^{self}(\{\mathbf{u}\}) + E^{dpl}(\{\mathbf{u}\}) + E^{dpl}(\{\mathbf{u}\}) + E^{short}(\{\mathbf{u}\}) \\
 & + E^{elas}(\{\eta\}) + E^{int}(\{\mathbf{u}\}, \{\eta\}),
 \end{aligned}
 \tag{2}$$

105 in which E^{self} is the self-energy of local-modes, E^{dpl} denotes the long-range dipole-dipole
 106 interaction, E^{short} is the short-range interaction of soft modes, E^{elas} denotes the contribution
 107 of elastic energy, and E^{int} is the interaction of local modes and strain. [An extensive overview](#)
 108 [and explanation on the energy terms of Equation 2 is provided in Ref. \[40\].](#)

109 The second term of Equation 1, whose energy is only related to AFD motions and their
 110 couplings with local modes and strains, is given by

$$\begin{aligned}
 E^{AFD} = & \sum_i [\kappa_A \omega_i^2 + \alpha_A \omega_i^4 + \gamma_A (\omega_{ix}^2 \omega_{iy}^2 + \omega_{iy}^2 \omega_{iz}^2 + \omega_{ix}^2 \omega_{iz}^2)] \\
 & + \sum_{ij} \sum_{\alpha\beta} D_{ij\alpha\beta} \omega_{i\alpha} \omega_{j\beta} \\
 & + \sum_i \sum_{\alpha\beta} C_{i\alpha\beta} \eta_l(i) \omega_{i\alpha} \omega_{i\beta} \\
 & + \sum_{ij} \sum_{\alpha\beta} K_{ij,\alpha\beta} u_{j,\alpha} \omega_{i,\alpha} \omega_{j,\beta} \\
 & + \sum_i \sum_{\alpha\beta\gamma\delta} E_{\alpha\beta\gamma\delta} \omega_{i\alpha} \omega_{i\beta} u_{i\gamma} u_{i\delta},
 \end{aligned}
 \tag{3}$$

111 where indices i and j numerate the lattice sites and α , γ , ν , and δ are the Cartesian
 112 components. In Equation 3, the first term shows the contribution of self-energy of the tilting
 113 of the oxygen octahedra while the second term shows the contribution of the short range
 114 interactions of AFD motions. The third term shows the coupling of the AFD motions with
 115 strain. The fourth and fifth terms show the contributions of trilinear and biquadratic energies
 116 coupling the local modes and AFD distortions. More details about AFD energy contributions
 117 can be found in Ref. [41]. The third term of Equation 1 shows the magnetic moments and
 118 the coupling of local modes, strain, and antiferrodistortive motions with magnetic moments

119 as given in the following

$$\begin{aligned}
E^{MAG} = & \sum_{ij\alpha\gamma} Q_{ij\alpha\gamma} m_{i\alpha} m_{j\gamma} \\
& + \sum_{ij\alpha\gamma} S_{ij\alpha\gamma} m_{i\alpha} m_{j\gamma} \\
& + \sum_{ij,\alpha\gamma\nu\delta} E_{ij,\alpha\gamma\nu\delta} m_{i\alpha} m_{j\gamma} u_{i\nu} u_{j\delta} \\
& + \sum_{ij,\alpha\gamma\nu\delta} F_{ij,\alpha\gamma\nu\delta} m_{i\alpha} m_{j\gamma} \omega_{i\nu} \omega_{j\delta} \\
& + \sum_{ijl,\alpha\gamma} G_{ijl,\alpha\gamma} \eta_l(i) m_{i\alpha} m_{j\gamma} \\
& + \sum_{ij} L_{ij} (\boldsymbol{\omega}_i - \boldsymbol{\omega}_j) \cdot (\mathbf{m}_i \times \mathbf{m}_j).
\end{aligned} \tag{4}$$

120 In Equation 4, the first term indicates the dipolar interactions of the magnetic moments.
121 The second term shows the short-range magnetic exchange coupling. The third, fourth, and
122 fifth terms indicate the contribution of local modes, AFD motions, and strain in magnetic
123 exchange interaction, respectively. The sixth term shows the contribution of the tilting of
124 oxygen octahedra through Dzyaloshinskii-Moriya interaction [42–44]. In addition to the
125 energy contributions discussed above, if external magnetic and/or electric fields are applied,
126 their contribution to the energy as $-\sum_i \mathbf{m}_i \cdot \mathbf{H}$ for magnetic field and $-\sum_i \mathbf{P}_i \cdot \mathbf{E}$ for
127 electric field should be added to the total energy terms. The electrical polarization, \mathbf{P} ,
128 can be computed from its proportionality to the local modes via $\mathbf{P} = \frac{Z^* \mathbf{u}}{V_0}$ in which Z^*
129 is Born effective charge obtained from first-principle calculations and its value for BFO is
130 $Z^* = 5.68 C$ [45], while V_0 is the volume of the unit cell.

131 To predict the dynamical properties of $BiFeO_3$ within the Effective Hamiltonian frame-
132 work used here, Newtonian equations of motion are applied for the ionic degrees of freedom
133 (i.e. local modes, AFD, strains) discussed in Equation 1 where the forces were computed by
134 taking partial differential of the total energy terms similar to the techniques used in Refs.
135 [46–48]. Moreover, as demonstrated in Ref. [13], the dynamics of the magnetic moments
136 are considered through the Landau–Lifshitz–Gilbert (LLG) equation [49] as indicated in the
137 following:

$$\begin{aligned}
\frac{d\mathbf{m}_i}{dt} = & -\Gamma \mathbf{m}_i \times [\mathbf{B}_{\text{eff}}^i(t) + \mathbf{b}_{\text{fl}}^i(t)] - \Gamma \frac{\lambda}{|\mathbf{m}_i| (1 + \lambda^2)} \mathbf{m}_i \\
& \times \{ \mathbf{m}_i \times [\mathbf{B}_{\text{eff}}^i(t) + \mathbf{b}_{\text{fl}}^i(t)] \}.
\end{aligned} \tag{5}$$

138 Here, $\mathbf{B}_{\text{eff}}^i$ is the effective magnetic field which acts on the i th magnetic moment and is
 139 given by $\mathbf{B}_{\text{eff}}^i = -\partial E_{\text{tot}}/\partial \mathbf{m}_i$, while Γ and λ are gyromagnetic and damping coefficients,
 140 respectively. \mathbf{b}_{fl}^i is a fluctuation field which is also acting on the i th magnetic moment.
 141 More details on the dynamics of magnetic properties calculations are given in Ref [13].

142 III. RESULTS AND DISCUSSION

143 A. Polarization, Magnetization, and Magnetoelectric Coefficient

144 Here and as shown in Figure 1(a), we investigate BFO with two domains separated by
 145 a 109° domain wall through a periodic $20 \times 10 \times 10$ supercell (For comparison, Figure 1(b)
 146 displays a monodomain). We have investigated 71° and 180° domain configurations as well,
 147 and the results of our analyses are consistent with 109° configuration. However, since 109°
 148 domain walls are the ones that are found most frequently in BFO bulk materials and the
 149 most seen ferroelastic domain walls in single crystal samples of BFO, the results for 109°
 150 configuration are shown and discussed here [50, 51]. In Figure 1(a), the left half-side along
 151 the x-axis (which lies along the pseudo-cubic [100] direction), is made of electric dipoles
 152 being initially along [111] while the right half-side is formed by electric dipoles initially
 153 lying along $[\bar{1}\bar{1}\bar{1}]$. This supercell is subjected to a magnetic field, $H = H_{dc} + h_{ac} \sin(\omega t)$,
 154 that is applied along the $[\bar{2}11]$ direction and which is a combination of a dc field with an
 155 ac field of ω frequency. We chose $H_{dc} = 245 \text{ T}$ and $h_{ac} = 61.2 \text{ T}$ in order to get good
 156 statistics on the quadratic ME coefficients since, for such fields, linear magnetoelectricity
 157 has a small effect with respect to the quadratic magnetoelectric coefficients in BFO (see
 158 Refs. [36, 38] and the Supplementary Information of Ref. [26]). However, and as shown
 159 in the Supplementary Material, the simulations were performed at lower magnetic fields to
 160 make sure that the observed results are still valid at magnetic fields that can be applied in
 161 practical applications.

162 Note that, in the entire supercell, the initial magnetization is along the $[\bar{2}11]$ direction,
 163 i.e., along which the magnetic field is applied, while the G-type antiferromagnetic vector is
 164 along the perpendicular $[01\bar{1}]$ direction. Furthermore, ω is fixed during each simulation, but
 165 is allowed to vary from 10 GHz to 1300 GHz between different computations. The upper
 166 limit of investigated frequencies is intentionally chosen such as to avoid resonating with the

167 A_1 phonon frequencies (known to occur at much higher frequencies of the order of ~ 4300
 168 and ~ 7000 GHz [25, 52]), but rather to focus on the possibility of having electromagnons
 169 associated with domain walls and thus occurring at lower frequencies (note that we do see
 170 these A_1 phonon frequencies in our simulations but do not report them here).

171 Note also that the homogeneous strain has been recently predicted to combine with optical
 172 phonons and magnons to create a new quasi-particle, coined electroacoustic magnon, that
 173 can result in the emergence of resonance in the magnetoelectric response [26]. Since we
 174 are especially interested here in revealing domain-wall-induced electromagnons (i.e., mixing
 175 of optical vibrations with magnons in a multidomain structure), we perform two kinds
 176 of MD simulations: one for which the homogeneous strain is clamped (implying that no
 177 electroacoustic magnon can exist) *versus* another for which such strain is fully relaxed. The
 178 inhomogeneous strain, on the other hand, is considered in the effective Hamiltonian and can
 179 vary during the course of all types of simulations conducted here.

180 Note also that, in multiferroics, the electrical polarization under the influence of applied
 181 electric and magnetic fields can be obtained by differentiating the expansion of the free
 182 energy with respect to electric field [5]. In the absence of external applied electric fields
 183 and when neglecting the linear magnetoelectric coefficient, the temporal evolution of the
 184 polarization as a function of our applied magnetic fields is given by

$$P(t) = P_{(t=0)} + \beta(0, \omega) H_{dc} h_{ac} \sin(\omega t) + \beta(\omega, \omega) \frac{1}{2} h_{ac}^2 \sin^2(\omega t). \quad (6)$$

185 where $\beta(0, \omega)$, on which we primarily focus, is the quadratic ME coefficient coupling the ac
 186 and dc fields, while $\beta(\omega, \omega)$ couples the ac field with itself and generates a second harmonic
 187 at 2ω .

188 The real and imaginary parts of $\beta(0, \omega)$ can thus be obtained by

$$\beta'(0, \omega) = \frac{\frac{2}{\tau} \int_0^\tau (P - P_{(t=0)}) \sin(\omega t) dt}{H_{dc} h_{ac}}, \quad (7)$$

189 and

$$\beta''(0, \omega) = \frac{\frac{2}{\tau} \int_0^\tau (P - P_{(t=0)}) \cos(\omega t) dt}{H_{dc} h_{ac}}, \quad (8)$$

190 in which τ is the overall time of the simulation. More details about the derivation of
 191 equations (7) and (8) are provided within the Supplementary Material.

192 **B. Frequency Analysis**

193 Let us first consider the case of the aforementioned multidomain structure *when the homo-*
 194 *geneous strain is clamped*. During and at the end of the simulation, the overall polarization
 195 is along the [100] direction, since the y- and z-components of the dipoles in the left and right
 196 domains cancel each other. Furthermore, the direction of the magnetic moments during and
 197 at the end of the simulation is found to be along the applied *dc* magnetic field, i.e., $[\bar{2}11]$, in
 198 both domains.

199 We then perform Fourier analysis of the temporal evolution of this electrical polarization
 200 and resulting magnetization along different directions (in order to, e.g., reveal the directions
 201 of vibration of phonons and magnons that are dynamically coupled to form electromagnons).
 202 Note that here we are dealing with two types of frequencies, one being the frequency of
 203 applied magnetic field which is denoted by ω , while the other is the frequency spectrum
 204 that we get as a result of Fourier analyses of the dynamical evolution of the polarization or
 205 magnetization under the applied field. For instance, the frequency spectra of the electrical
 206 [100]-component of the dipoles (direction of polarization, as well as normal to the ferroelectric
 207 domain wall) and the perpendicular, [011]-component, when the frequency of the applied
 208 magnetic field, ω , is 10 GHz are demonstrated by red solid lines in Figures 2a and 2c,
 209 respectively, and up to about 1300 GHz. Apart from the frequency of applied field and its
 210 second harmonic at 10 and 20 GHz, two peaks are observed at ~ 390 and ~ 1150 GHz in the
 211 spectrum of the [100]-component (Figure 2a). Also a peak is observed at ~ 700 GHz for the
 212 [011]-component (Figure 2c).

213 In addition, the Fourier analyses of the magnetization were performed for the $[\bar{2}11]$ -
 214 component which is also the direction of the magnetic moments and the applied magnetic
 215 field (Figure 2b) as well as the $[01\bar{1}]$ -component which is also a direction that is perpen-
 216 dicular to both polarization and magnetization (Figure 2d). Since the peaks at ~ 390 GHz
 217 and ~ 1150 GHz are present in the Fourier analyses of both [100]-component of polarization
 218 and $[\bar{2}11]$ -component of magnetization, these peaks represent *electromagnons* for which the
 219 optical phonon has a component of its vibration along the direction that is perpendicular to
 220 the domain wall and the magnon has a component of its vibration along the magnetization.
 221 (It should be noted that, we found such electromagnons for the 71° domain configuration at
 222 ~ 455 GHz and ~ 1390 GHz. Also, for the 180° domain configuration, such electromagnons

223 are found to be at 240 GHz and 1430 GHz). Similarly, the peaks at ~ 700 GHz seen in Figs
 224 2c and 2d characterize an electromagnon made of an optical phonon that has a vibration
 225 direction with a component lying along the $[011]$ direction and of a magnon for which the
 226 vibration has a finite component along $[01\bar{1}]$ (note that $[011]$ and $[01\bar{1}]$ are both lying in the
 227 plane of the domain wall but are perpendicular to each other). Interestingly, and as verified
 228 by black solid lines in Figures 2a to 2d, these three peaks disappear when the simulations
 229 are performed for the case of monodomain structure (whether the homogeneous strain is
 230 clamped or unclamped). In other words, one can conclude that the 390, 1150, and 700 GHz
 231 modes are *domain-wall-induced* electromagnons (to be denoted as DWI electromagnons in
 232 the following). Such observations are also consistent with previous findings that vibrations
 233 of the order of 360, 700 and ~ 1200 -1800 GHz caused by domain wall motions were pre-
 234 dicted for the case of BFO adopting domain structures [50, 53] (note that other domain wall
 235 activities, but with lower frequencies, were also reported in Refs. [54, 55] but the limitation
 236 of molecular dynamics methods below the range of few GHz prevents us from determining
 237 them).

238 Figures 2a, 2b, 2c, and, 2d also report the case of a multidomain but *for unclamped homoge-*
 239 *neous strain*, which is represented by dashed blue lines. In this latter case, three additional
 240 peaks are observed at 70, 100 and 270 GHz with respect to the case of clamped multidomain,
 241 for both the Fourier transform of the temporal evolution of the polarization and magnetiza-
 242 tion. These three peaks, therefore, indicate that strain can mix with optical lattice vibrations
 243 and magnons to produce new particles – which is in-line with the recent prediction of the
 244 so-called electroacoustic magnons for unclamped monodomain in Ref. [25]. Note that these
 245 electroacoustic magnons at 70 and 270 GHz in our multidomain involve vibration of the
 246 polarization along the normal of the domain wall (see Fig. 2a) while that at 100 GHz is
 247 better seen in the oscillation of the electric dipoles perpendicularly to that normal (cf Fig.
 248 2c). Moreover, the magnon associated with these electroacoustic magnons at 70, 100 and
 249 270 GHz have vibrations along both $[\bar{2}11]$ (that are stronger) and $[01\bar{1}]$ (that are weaker),
 250 as demonstrated by Figures 2b and 2d.

251 C. Magnetolectric Response

252 Let us from now on focus on results obtained *for the case of unclamped homogeneous strain*
 253 in the multidomain structure, since we now know the origins of all the peaks appearing
 254 in Figure 2. The real and imaginary parts of the quadratic magnetolectric coefficient,
 255 $\beta(0, \omega)$, for polarization lying along the [100] and [011] directions, are then computed using
 256 Equations (7) and (8) at each frequency of applied magnetic field from 10 to 1300 GHz in
 257 that case. They are shown in Figures 3a and 3b, respectively. The calculated values are
 258 in agreement with experimental measurements of β of bulk BFO at small frequencies. For
 259 instance, the obtained value for the real part of $\beta(0, \omega)$ along the [100] direction by our MD
 260 simulations when the frequency of applied magnetic field is 20 GHz and the homogeneous
 261 strain is unclamped, is $2.457 \times 10^{-8} C/m^2T^2 = 0.388 \times 10^{-19} s/A$ which remarkably agrees
 262 with the observation of $\beta_{311} = 0.3 \times 10^{-19} s/A$ reported in Reference [56] for the static case.
 263 Also, in measurements that were performed in high magnetic fields [57], the quadratic ME
 264 coefficient was found to be $\beta_{122} = \beta_{212} = \beta_{221} = -\beta_{111} = -5 \times 10^{-9} C/m^2T^2$ which is in
 265 reasonable agreement with the value of $-1.004 \times 10^{-9} C/m^2T^2$ that we have computed along
 266 the [011] direction at $\omega=20$ GHz. The ME coefficients exhibit resonances when the frequency
 267 of applied magnetic field, ω , approaches the frequency of the electroacoustic-magnon-like
 268 particles mentioned above, that are 70, 100 and 270 GHz here, as previously found for the
 269 case of a monodomain, for which electroacoustic magnons have resonant frequencies at 90
 270 and 267 GHz [25, 26].

271 Moreover, another important novelty of the present work is that resonances also happen
 272 when ω reaches the frequency of the three domain-wall-induced electromagnons. For in-
 273 stance, along the [011] direction, a nicely seen resonance takes place at $\omega=700$ GHz, while,
 274 along the [100] direction, one resonance is rather sharp at 390 GHz while the other one
 275 around 1150 GHz is broader and even seems to be made of several peaks – which is rem-
 276 iniscent of the broad response found in Ref. [53] around these frequencies for the domain
 277 wall motions. These results are also consistent with the dielectric and domain-wall-related
 278 modes A and B reported in Ref. [50] for which mode A around $12 \text{ cm}^{-1} \simeq 360$ GHz is
 279 sharp while mode B around $50 \text{ cm}^{-1} \simeq 1500$ GHz is broader. Our DWI electromagnons
 280 therefore provide a path to engineer devices based on the functionality of the domain walls.
 281 Although domain walls have already been employed in spintronics and for designing novel

282 devices [58–60], their presently discovered role in optimizing ME responses make them also
 283 fascinating for magnetoelectric-based devices. As a matter of fact, our results show that one
 284 can tune the frequency of the applied magnetic field with the frequency of the domain wall
 285 vibrations (DWI electromagnons) to maximize the ME response.

286 **D. Degree of localization of the domain-wall-induced electromagnons**

287 In order to investigate the degree of localization that these DWI electromagnons possess,
 288 a layer-by-layer analysis is performed at different planes that are parallel to the domain wall.
 289 As the dimension of the supercell is $20 \times 10 \times 10$, each 10×10 cross section of the supercell
 290 along the [100] direction is defined as a layer for which the average of magnetization and
 291 polarization can be computed from the MD simulation results. Also, the quadratic ME
 292 coefficient of each layer can be obtained via equations (7) and (8) but for which the overall
 293 polarization is replaced by the average polarization of each layer. The layers are indicated
 294 by their plane numbers, with the planes 10 and 20 corresponding to the domain wall layers
 295 whereas planes 5 and 15 correspond to the center of each domain. Since the magnetoelectric
 296 coefficients $\beta(0, \omega)$ are calculated at each frequency of applied magnetic field for frequencies
 297 up to 1300 GHz, in each plane that is parallel to the domain wall, the absolute value of the
 298 real part of magnetoelectric coefficient, $|Re(\beta(0, \omega))|$ (which is related to real-space location
 299 of optical phonons) is used to investigate the preferential location of the optical phonons.
 300 On the other hand, for each frequency of applied magnetic field, the Fourier analysis of
 301 magnetic moments provides a spectrum of the frequencies that are present in the vibration
 302 of the magnetic moments. Therefore, the Fourier intensity of the magnetization which is
 303 related to real-space location of magnons is used to investigate the preferential location of
 304 magnons. Here we used the Fourier analyses when the frequency of applied magnetic field
 305 is 10 GHz.

306 Figures 4a and 4b provide an example of the results on such analysis for the optical
 307 phonon (Figure 4a) and magnon (Figure 4b) associated with the frequency of 700 GHz.
 308 One can see that the maximum value of the resonance peak inside the domain (plane 5),
 309 which is $1.732 \times 10^{-8} C/m^2T^2$, jumps to $4.393 \times 10^{-8} C/m^2T^2$ at the domain wall (plane 10)
 310 showing that the ME coefficient is increased by 154%. Such comparison clarifies that not
 311 only the ME coefficient exhibits a resonance when the frequency of applied magnetic field is

312 700 GHz, but also the resonance is intensified at the domain walls. Furthermore, Figure 4b
 313 compares the intensity of Fourier analyses peaks around 700 GHz for magnetization along
 314 the $[01\bar{1}]$ direction in each layer. The Fourier intensity of vibrations of magnetization at
 315 $\omega=700$ GHz is stronger at the domain wall. Consequently, one can conclude that the DWI
 316 electromagnon at $\omega=700$ GHz can be seen as waves that adopt maximum at the domain
 317 wall, that is both the optical phonon and magnon forming such electromagnon preferen-
 318 tially localize at this ferroelectric domain wall. Such domain-wall-induced electromagnon's
 319 wave behavior and preferential localization are also found for the frequency of 1150 GHz
 320 (but for optical phonon vibrating along $[100]$ and magnons oscillating along $[\bar{2}11]$). As a
 321 matter of fact and as demonstrated in Figures 5(a) and (b), both the optical phonon and
 322 magnon of the domain-wall-induced electromagnon at 1150 GHz adopt wave-like behavior
 323 with maxima occurring in the vicinity of the domain walls (near plane 10). On the other
 324 hand, the DWI electromagnon having a resonant frequency of 390 GHz, while still possess-
 325 ing wave-like behavior, prefers to possess maxima occurring, both for its optical phonon and
 326 magnon, *near the middle of the domains* (near planes 5 and 15) rather than at the domain
 327 wall, as demonstrated in Figures 6a and 6b. Note that the Supplementary Material also
 328 reports information about localization/delocalization of the electroacoustic magnons and
 329 some results on inhomogeneous strain analysis.

330 E. The origin of the domain-wall-induced electromagnons

331 To have a clear understanding of the origin of the 390 GHz, 700 GHz and 1150 GHz
 332 electromagnons induced by the domain structure, we have analyzed the spectral energy
 333 density (SED) associated with local dipoles (Φ_P) and inhomogeneous stain (Φ_η) when the
 334 homogeneous strain was unclamped and for a $40\times 10\times 10$ supercell. In order to conserve the
 335 periodicity of the domain structure, the supercell is split into four alternating domains of
 336 equal width. Note that a larger supercell (i.e. $40\times 10\times 10$ as compared to $20\times 10\times 10$) is used
 337 to increase the wave-vector q resolution. Note also that Φ_X , with $X=P$ or η , describes the
 338 wave-vector \mathbf{q} and frequency ν resolved power of the kinetic energy spectrum for the vector
 339 degrees of freedom X . It is computed [61] as

$$\Phi_X(\mathbf{q}) = \frac{1}{N\tau} \sum_{\alpha=x,y,z} \left| \dot{X}_{\mathbf{q},\nu}^\alpha \right|^2, \quad (9)$$

340 where $\dot{X}_{\mathbf{q},\nu}^\alpha$ denotes the Fast Fourier Transform coefficient of the α component of the velocity
 341 \dot{X}^α . N and τ denote the total number of unit cells and the molecular dynamics trajectory
 342 time, respectively. The poles of Φ_P provide the spectrum of excitations independently of
 343 the mode's polarization vector. Specifically, Φ_P and Φ_η can be used to obtain dispersions of
 344 optical and acoustic phonons, respectively.

345 Practically, we use a single 0.5 ns molecular dynamics trajectory computed for this $N =$
 346 $40 \times 10 \times 10$ supercell. The integration time step is taken to be equal to 0.5 fs and the first
 347 10,000 steps of the trajectory are not taken into consideration. The velocities are sampled
 348 each 10 MD steps for a subsequent Fast Fourier transforms to the wave-vector and frequency
 349 domain.

350 Figure 7a and 7c shows the computed low-frequency spectra of polar excitations along
 351 the [100] pseudo-cubic direction (i.e., different panels on the left/right side correspond to
 352 different q-points along that direction) for mono- and multi-domain states, respectively. The
 353 spectrum for the mono-domain configuration (panel a of Fig. 7) clearly shows the dispersion
 354 of three electromagnon branches labeled (1)-(3). From the analysis of Φ_η , we also find that
 355 these branches follow the dispersion curves of acoustic phonons (not shown here). Branches
 356 (1) and (2) match the two transverse acoustic (TA) modes, while the third branch is mixed
 357 with the longitudinal acoustic (LA) mode.

358 Interestingly, the three described monodomain branches can be also clearly seen in the
 359 spectrum of the multidomain state (see panel c of Fig. 7). However, the multidomain
 360 spectrum additionally features multiple “satellite” peaks not present in the monodomain
 361 case (panel a of Fig. 7). These features most likely stem from the elastic scattering of the
 362 three mono-domain electromagnons by a periodic and static array of domain walls. In such
 363 hypothetical scenario, a single scattering event would conserve the mode frequency and lead
 364 to a shift of its momentum q_x by $\pm\Delta q_x$. The possible shifts

$$\Delta q_x = \frac{2}{20} \frac{\pi}{a_0}, \frac{6}{20} \frac{\pi}{a_0}, \frac{10}{20} \frac{\pi}{a_0} \quad (10)$$

365 correspond to the dominant Fourier components of the local mode distribution, or, in other
 366 words, are determined by the periodicity of the domain structure. Here a_0 indicates the
 367 5-atom lattice constant.

368 The simplest single scattering mechanism described above allows to explain the appear-
 369 ance of the predicted domain-wall-induced electromagnons at $q_x = 0$. Namely, the emergent

370 zone-center excitations at 390 GHz and 700 GHz can be matched with monodomain modes
 371 (1) and (3) at $q_x = \frac{2}{20} \frac{\pi}{a_0}$ that are scattered by $\Delta q_x = -\frac{2}{20} \frac{\pi}{a_0}$ (see panels a and b of Fig. 8),
 372 while the homogeneous 1150 GHz mode stems from scattering of mode (1) at $q_x = \frac{6}{20} \frac{\pi}{a_0}$ by
 373 $\Delta q_x = -\frac{6}{20} \frac{\pi}{a_0}$ (see panels a and c of Fig. 8).

374 Such hypothesis about the origin of the domain-wall-induced electromagnons is fur-
 375 ther supported by visualizing the local mode vibrations at $\nu_0 = 390$ GHz, 700 GHz and
 376 1150 GHz. To perform this analysis, we first note the deviation of the average local mode
 377 value $\delta \mathbf{u}(x, t) = \mathbf{u}(x, t) - \langle \mathbf{u}(x) \rangle$ within each x -layer of the supercell and at each MD step t
 378 from its time average $\langle \mathbf{u}(x) \rangle$. Then, we perform the time-frequency Fourier transformation
 379 $\delta \mathbf{u}(x, t) \rightarrow \delta \mathbf{u}(x, \nu)$ that allows us to look at how the layer-averaged local modes change
 380 along the [100] direction and only at the frequencies of interest (i.e., at these ν_0). We per-
 381 form the described procedure using the generated MD trajectories for the multidomain case
 382 which technically corresponds to isolating the excitations pattern at a specific frequency
 383 in Figure 7c. Figure 9 shows how the resulting complex amplitudes $\delta u_\alpha(x, \nu_0)$ and phases
 384 $\theta_\alpha = \arg(\delta \mathbf{u}_\alpha(x, \nu_0))$ change with x for each Cartesian component α of the local modes.

385 As expected from the scattering picture, the amplitudes of local modes displacements at
 386 $\nu_0 = 390$ GHz (Fig. 9a) and $\nu_0 = 700$ GHz (Fig. 9b) have a pronounced $q_x = \frac{2}{20} \frac{\pi}{a_0}$ character
 387 with half of the wave period falling within each of the domains (Figure 7b). At the same
 388 time, and as also consistent with Figure 8c, the amplitudes $\delta u_\alpha(x, \nu_0)$ at $\nu_0 = 1150$ GHz
 389 (see Figure 9c) have a different, three half-period, character rather described by $q_x = \frac{6}{20} \frac{\pi}{a_0}$
 390 (Figure 7b). This latter periodicity is more clearly visible in the dependence of the phase
 391 on x at $\nu_0 = 1150$ GHz (see Figure 9f).

392 It is also interesting to note the markedly distinct behavior of the phase plots at
 393 $\nu_0 = 390$ GHz and $\nu_0 = 1150$ GHz from the complex phase distribution at $\nu_0 = 700$ GHz.
 394 Indeed, the two former feature a constant (flat) phase $\theta_\alpha(x)$ dependencies on x within each
 395 domain (Figures 9d and f). Such distribution along with a spatially modulated amplitudes
 396 (Figures 9a and c) are unique signatures of standing waves. Notably, the standing-wave
 397 character (confinement) of these modes also goes in-line with the TA nature of the original
 398 monodomain branch (1) giving rise to these modes. Indeed, similar situations were pre-
 399 viously reported in references [62, 63] describing the confinement of TA acoustic phonons
 400 in semiconductor superlattices. In contrast, the $\nu_0 = 700$ GHz vibration pattern rather cor-
 401 responds to a propagating wave as the latter typically yield a linear phase dependence on

402 the coordinate (see Figure 9 e). This observation also goes in-line with our prediction of
 403 the common origin of the $\nu_0 = 390$ GHz and $\nu_0 = 1150$ GHz electromagnons distinct from
 404 that of the $\nu_0 = 700$ GHz domain-wall-induced mode. Finally, from Figures 9 it can be
 405 further noted that for all of the considered frequencies the vibration patterns are neither the
 406 classical standing nor classical propagating waves. Indeed, the amplitude plots in Figures 9
 407 clearly indicate that all modes are mixed with Gamma point vibrations while the amplitude
 408 deviations at the domain walls indicate the presence of domain-wall localized modes. More-
 409 over, the phase velocity of the propagating electromagnon at $\nu_0 = 700$ GHz clearly increases
 410 upon approaching the domain walls (Figure 9e) while, at the same time, its amplitude
 411 experiences a drop. Such change of the phase velocity can be equivalently described as a
 412 mixing of the propagating mode with a domain-wall localized mode and a standing wave
 413 at $q_x = \frac{2}{20} \frac{\pi}{a_0}$. All of the described deviations are due to the aforementioned scattering
 414 which couples phonons with wavevectors differing by the multiples of $\frac{2}{20} \frac{\pi}{a_0}$ due to which all
 415 of the three modes have a mixed character. To complete the picture, we summarize the
 416 information presented in Figure 9 by plotting the instantaneous snapshots of the calculated
 417 profiles of dipole vibrations at $\nu_0 = 390$ GHz, 700 GHz and 1150 GHz in Figure 10.

418 Based on the gathered insights, we now fit (as an example) the values of the Fourier
 419 intensity peaks of ME coefficient, $\beta(0, \omega)$, along the [100] direction and at 390 GHz by a
 420 function of the form

$$y = A + C|\sin(q(x - x_0))| \quad (11)$$

421 where $q = \frac{2}{20} \frac{\pi}{a_0}$ is the scattering vector for that frequency, A is the amplitude of the zone-
 422 center mode and C can be seen as an x-component amplitude of the confined electromagnon
 423 at q . One can clearly see that such function fits well the data of Fig. 6a, which confirms
 424 our proposed concept of scattering. Similarly, the values of the Fourier intensity peaks of
 425 magnetization along the $[\bar{2}11]$ direction at 390 GHz were fitted by a function of the form

$$y = A + C'|\sin(q(x - x_0))| + D'\sin(q(x - x_0)) \quad (12)$$

426 where q is still equal to $\frac{2}{20} \frac{\pi}{a_0}$, C' is related to the x-component contribution of the elec-
 427 tromagnon as in Eq.(11), while D' originates from its y- and z-components. Once again,
 428 this function fits well the data of Fig. 6b, which is another confirmation that scattering is
 429 the main source of the domain-wall-induced electromagnons. It should be noted that the
 430 difference between the data and the fit at domain walls comes from the modes localized on

431 the domain walls.

432 **IV. SUMMARY**

433 In summary, we report here the existence and explain the origin of domain-wall-induced
434 electromagnons, and discover that they enhance the magnetoelectric response of the mate-
435 rial to the application of external magnetic fields. Obtaining such results for bisumth ferrite
436 not only implies that ME enhancement can be achieved at room temperature, but also
437 infers that such enhancement can be generalized to other multiferroics possessing multido-
438 mains. Interestingly, the optical phonon and magnons forming such domain-wall-induced
439 electromagnons behave as waves for which the maxima jointly occur in some specific parts
440 of the multidomain, namely near the domain walls for the 700 and 1150 GHz frequencies
441 *versus* near the middle of each domain for the 390 GHz frequency. The presently discov-
442 ered domain-wall-induced electromagnons may thus be of some advantages for, e.g., the
443 design of (i) novel ultra-fast energy-efficient data storage devices via local probes and (ii)
444 and of electrically-driven configurable magnonic circuits taking advantage of the spatial
445 preferential localization of both the phonons and magnons making the domain-wall-induced
446 electromagnons.

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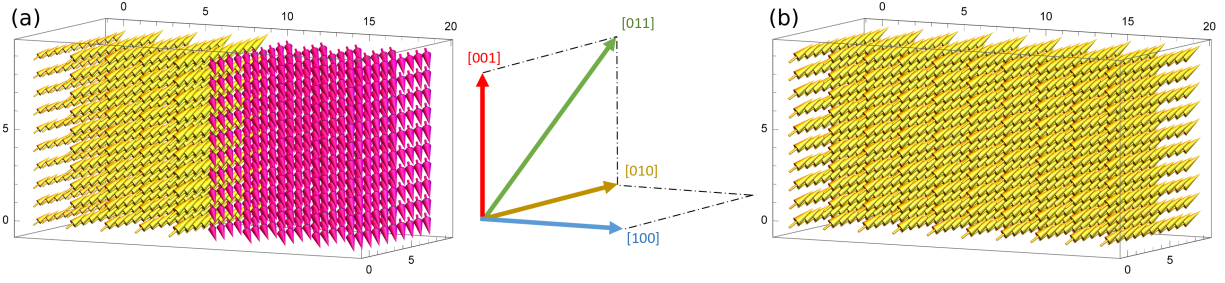


FIG. 1: (Color online) Schematic representation of the electric dipole moments for a $20 \times 10 \times 10$ supercell of BiFeO_3

- (a) a multidomain structure in which electric dipole moments are initially along the $[111]$ direction in the first (left) domain and along $[\bar{1}\bar{1}\bar{1}]$ in the second (right) domain; (b) a monodomain structure in which all electric dipole moments are along the pseudocubic $[111]$ direction

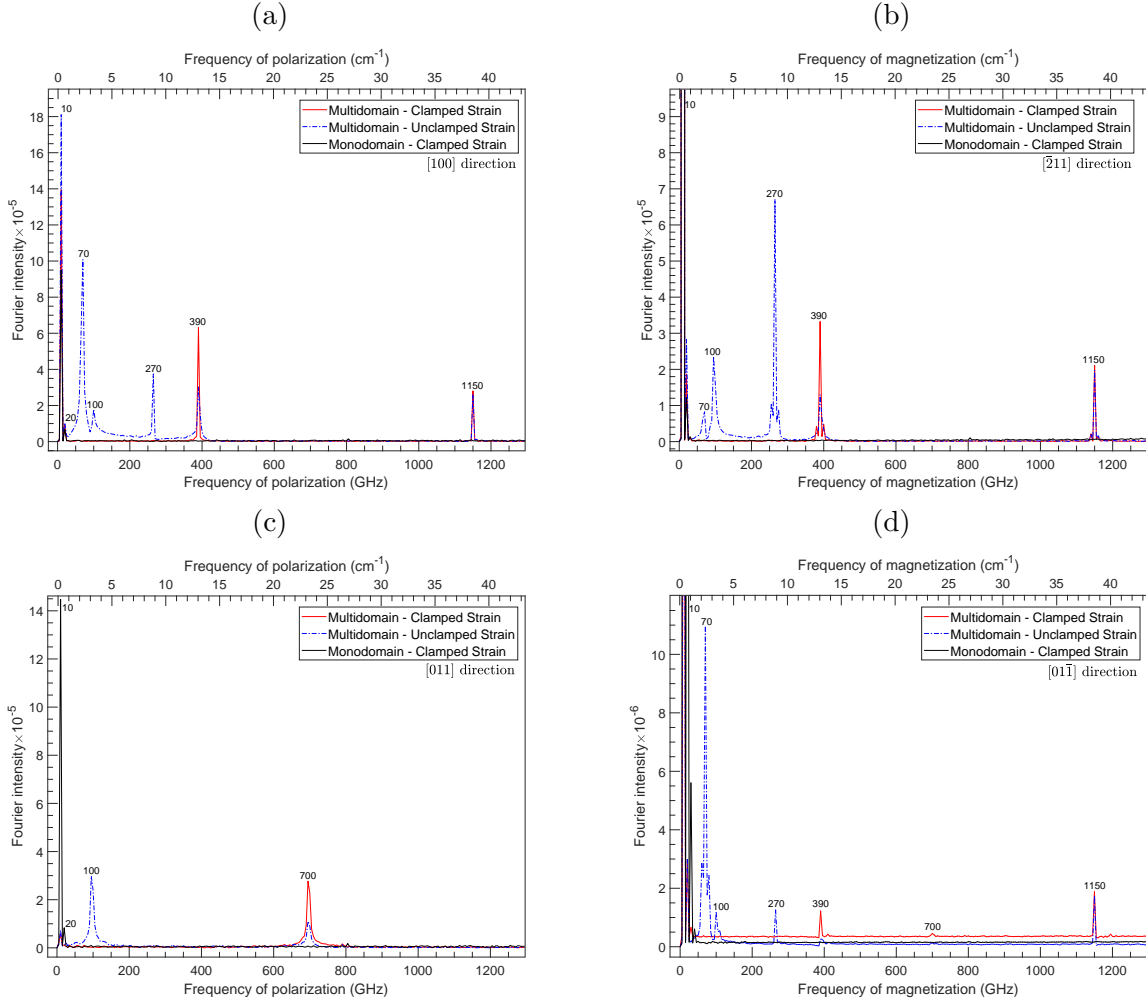


FIG. 2: (Color online) The Fourier analyses

The Fourier analyses of the polarization (panel a is for polarization along the $[100]$ direction while panel c is for polarization along the $[011]$ direction) and magnetization (panel b is for magnetization along the $[\bar{2}11]$ direction while panel d is for magnetization along the $[01\bar{1}]$ direction) for the case of monodomain when the strain is unclamped (black) and for the case of multidomain when the homogeneous strain is unclamped (blue) and clamped (red). Here, the frequency of the applied magnetic field is 10 GHz. Note the change of vertical scale between

Panels (b) and (d)

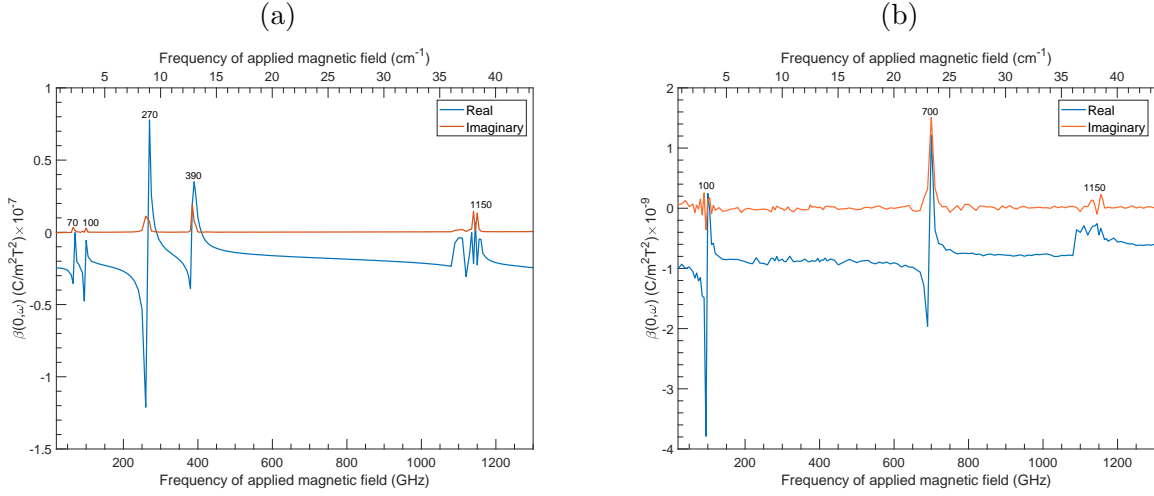


FIG. 3: (Color online) Frequency dependence of the quadratic magnetoelectric coefficient $\beta(0, \omega)$ along the [100] direction (panel a) and the [011] direction (panel b) when the homogeneous strain is unclamped. Note the change of vertical scale between Panels (a) and (b).

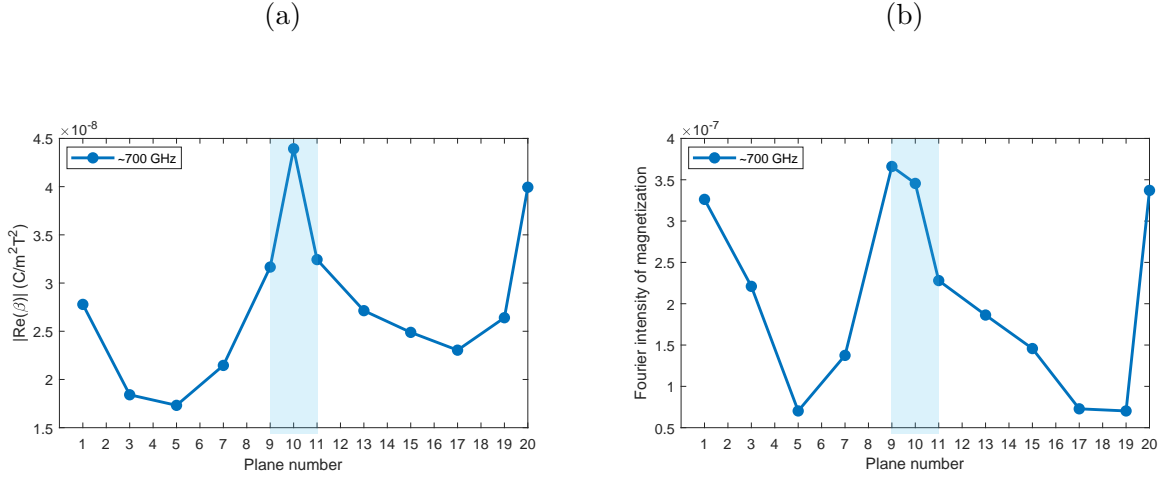


FIG. 4: (Color online) The layer-by-layer analysis for the domain-wall-induced electromagnons at 700 GHz

Layer-by-layer comparison of the intensity of the (a) resonance peaks of the real part of ME coefficient, β , along the $[011]$ direction when the frequency of applied magnetic field is 700 GHz; (b) Fourier peaks around 700 GHz for magnetization vibrations along the $[01\bar{1}]$ direction; For panels (b) the frequency of applied magnetic field is 10 GHz.

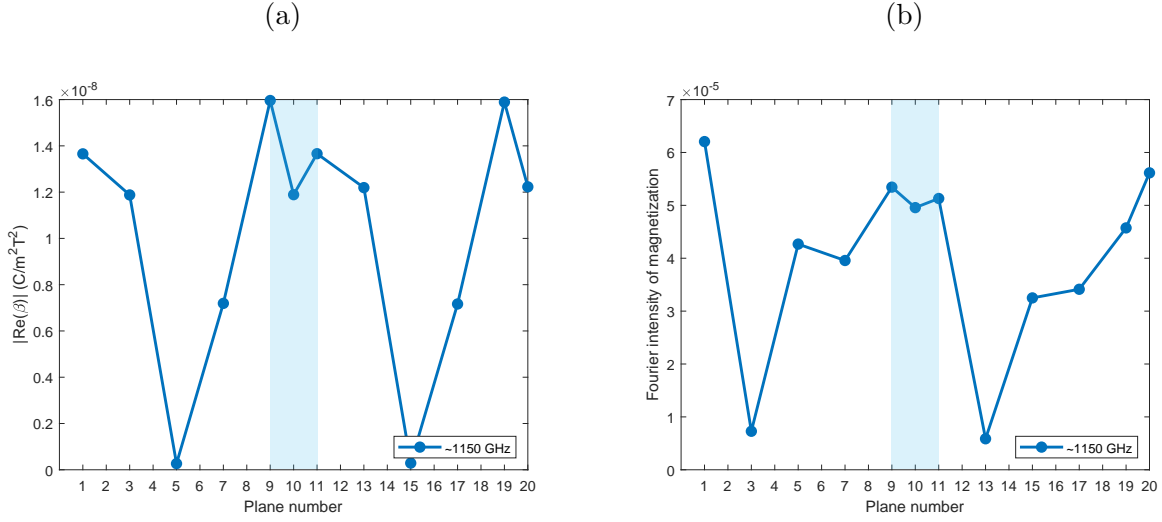


FIG. 5: (Color online) The layer-by-layer analysis for the domain-wall-induced electromagnons at 1150 GHz

Panel (a) shows the magnetolectric coefficient, β , along the $[100]$ direction when the frequency of applied magnetic field is 1150 GHz; Panels (b) shows the Fourier spectrum of magnetization, along the $[\bar{2}11]$ direction for 1150 GHz. For panels (b) the frequency of applied magnetic field is 10 GHz.

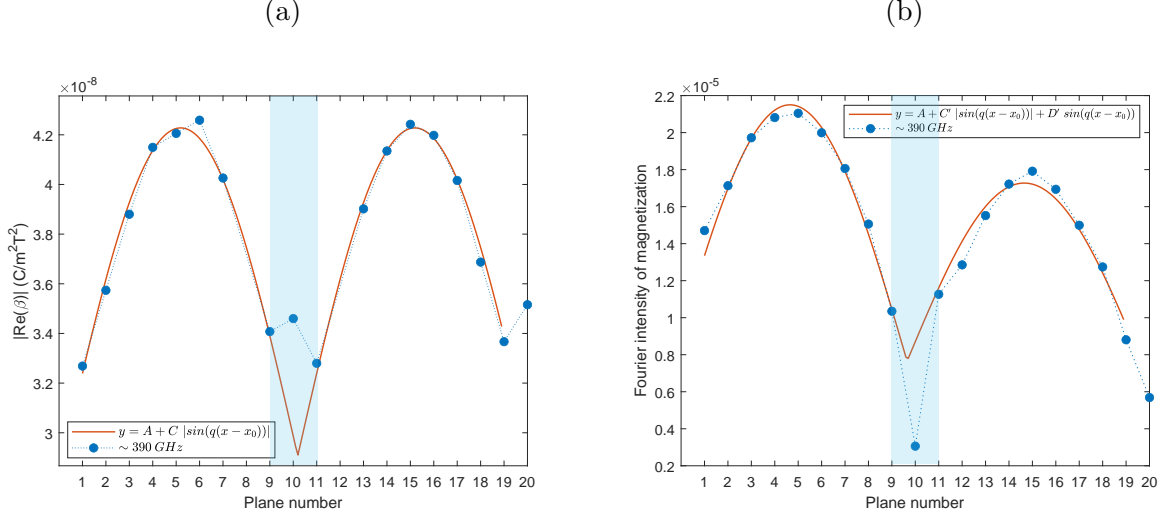


FIG. 6: (Color online) The layer-by-layer analysis for the domain-wall-induced electromagnons at 390 GHz

Layer-by-layer comparison of the intensity of the (a) resonance peaks of the real part of ME coefficient, β , along the $[100]$ direction when the frequency of applied magnetic field is 390 GHz; (b) Fourier peaks around 390 GHz for magnetization vibrations along the $[\bar{2}11]$ direction. For panel(b) the frequency of applied magnetic field is 10 GHz.

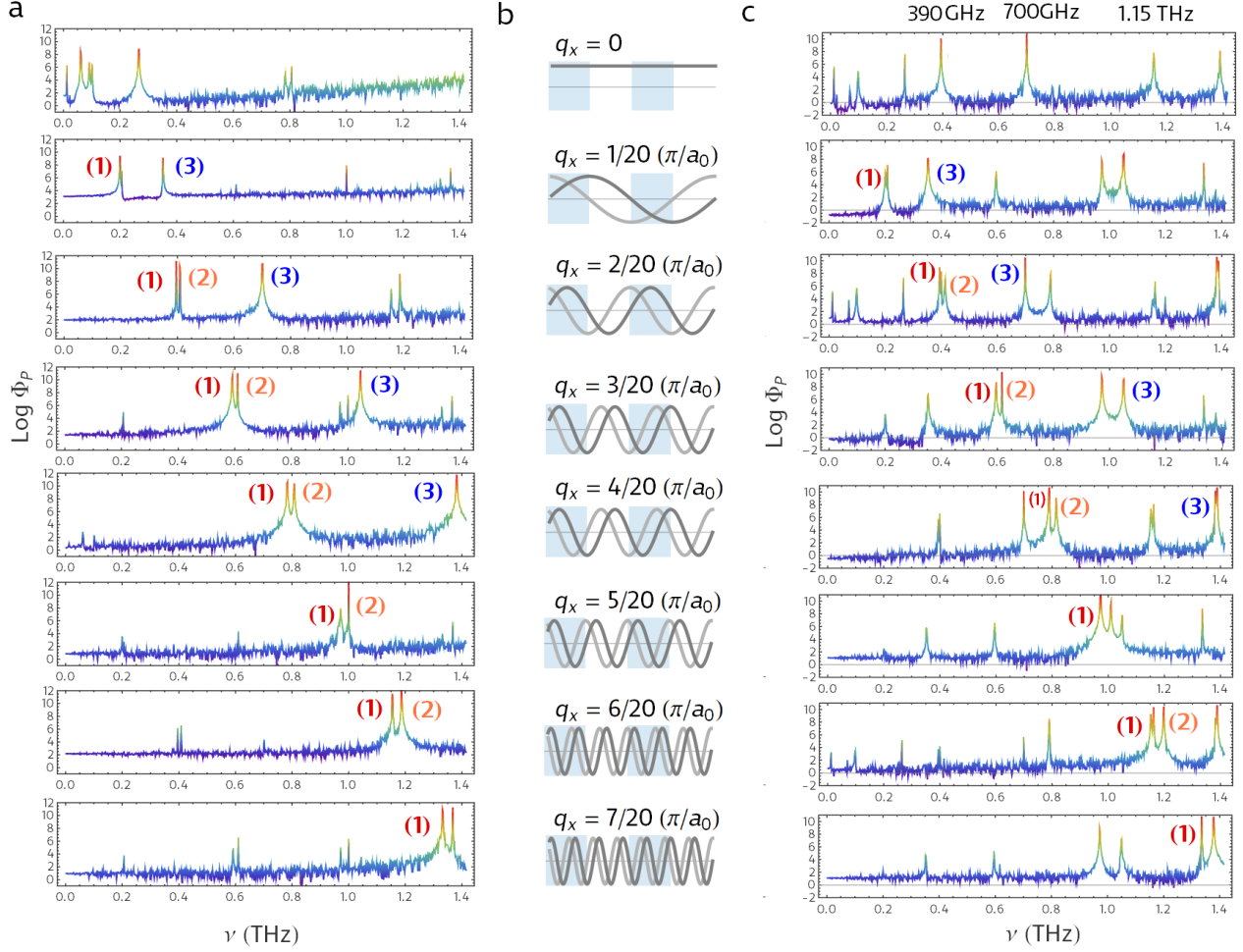


FIG. 7: (Color online) Low-frequency spectrum of polar excitations for monodomain and multidomain states.

Each sub-panel in panels (a) and (c) shows the dependence of the logarithm of the spectral energy density Φ_P on frequency at a single wavevectors $\mathbf{q} = (q_x, 0, 0)$. The labels (1), (2) and (3) indicate the relevant electromagnon branches. The value of q_x for each row of sub-panels is indicated in panel (b). Panel (b) also contains a schematic representation of a $40 \times 10 \times 10$ supercell with four alternating domains (blue and white regions) along with examples of the \mathbf{q} wave modulations.

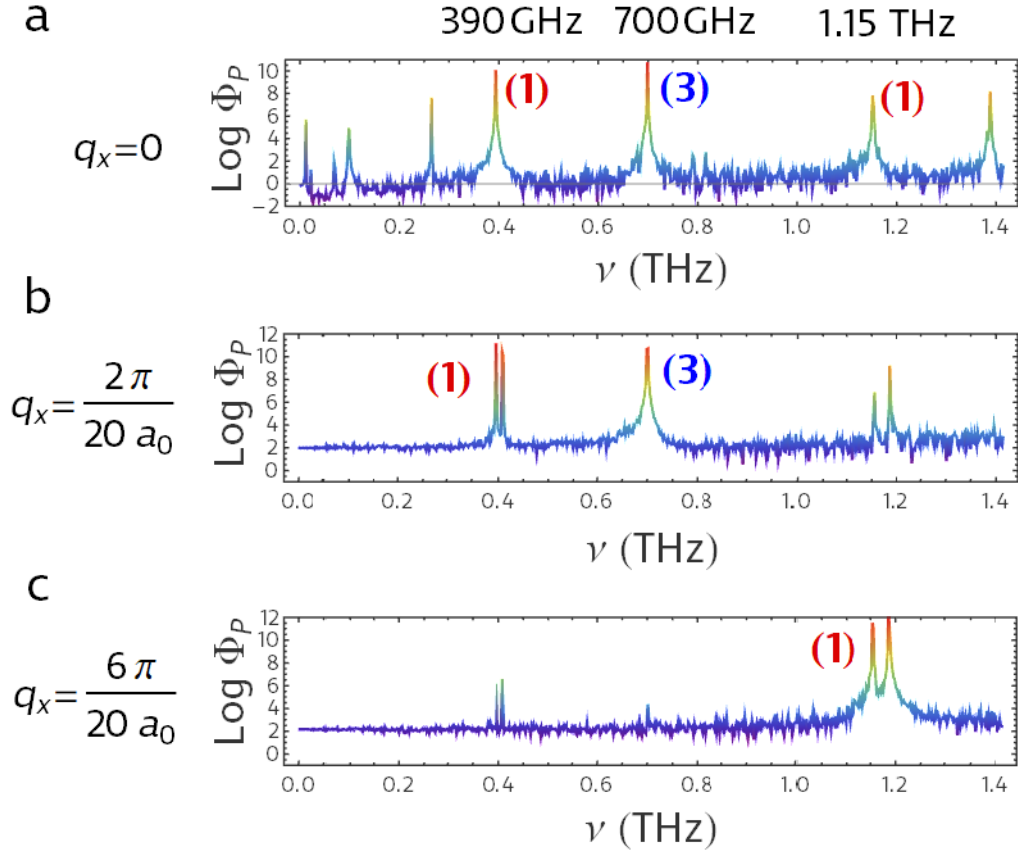


FIG. 8: (Color online) Origin of the zone-center electromagnons.

Panel (a) shows the logarithm of the optical phonon SED at $q_x = 0$ for the multidomain state.

Panels (b) and (c) shows the logarithm of the optical phonon SED for the monodomain state at

$q_x = \frac{2}{20} \frac{\pi}{a_0}$ and $q_x = \frac{6}{20} \frac{\pi}{a_0}$, respectively. Labels (1) and (3) indicate the matching frequencies of

the mode before (panels b,c) and after a single scattering event (panel a).

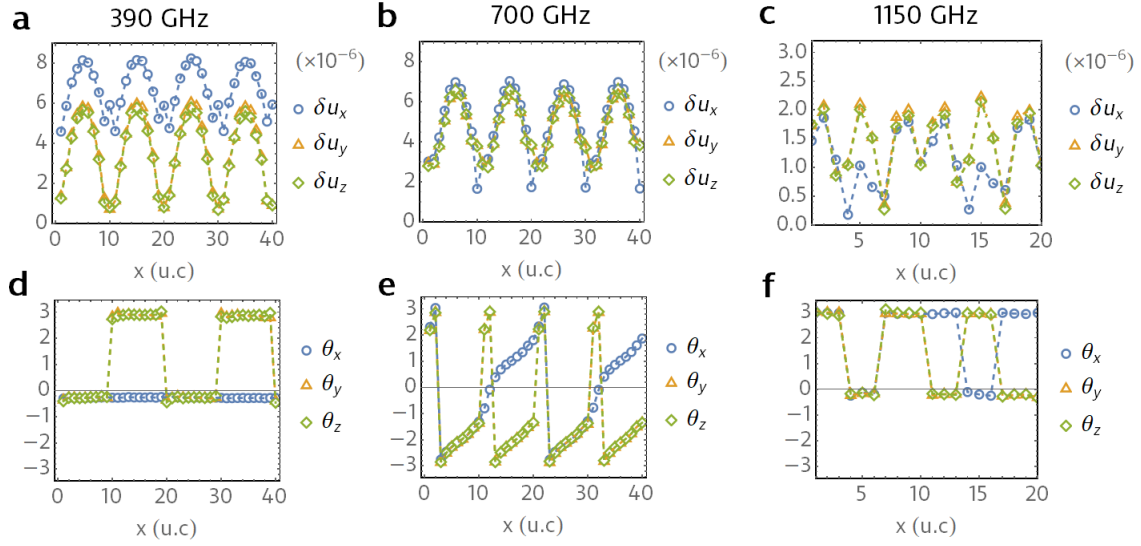


FIG. 9: (Color online) Analysis of local mode vibrations at emergent electromagnon frequencies.

Panels a, b and c (d, e and f) show the changes of the complex amplitude (phase) of the local mode vibrations along the [100] direction at 390 GHz, 700 GHz and 1150 GHz, respectively.

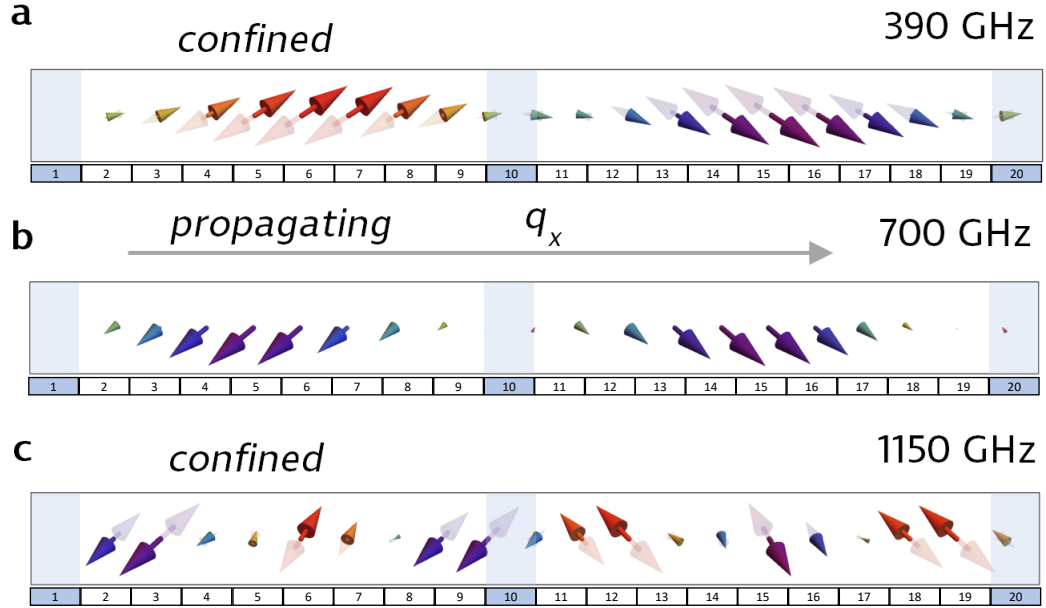


FIG. 10: (Color online) Instantaneous distribution of the local mode deviation vectors for the three considered domain-wall-induced electromagnons.

The arrows indicate the local mode deviations with red (purple) color corresponding to vector with positive (negative) z components. Electromagnons at 390 GHz and 1150 GHz originate from the same monodomain electromagnon branch and have predominantly confined (standing-wave) character. The 700 GHz domain-wall-induced mode features a pronounced propagating wave nature.