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1 **Electric-field-control of electromagnons' frequency in multiferroics**

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Abstract

13

14 Electromagnons, which are coupled polar and magnetic excitations in magnetoelectric materials,
15 are of large interest for electronic and computing technological devices. Using Molecular Dynamics
16 simulations based on an *ab-initio* effective Hamiltonian, we predict that the frequency of several
17 electromagnons can be tuned by the application of electric fields in the model multiferroic BiFeO₃,
18 with this frequency either increasing or decreasing depending on the selected electromagnon. In
19 particular, we show that the frequency of electromagnons localized at ferroelectric domain walls can
20 be tuned over a 200 GHz range by realistic dc electric fields. We interpret the realized frequency
21 increase (respectively, decrease) by local hardening (respectively, softening) of the associated polar
22 phonons which couples to the applied electric field. The increase *versus* decrease of the elec-
23 tromagnons' frequency is further found to be correlated with the real-space localization of such
24 phonons.

25 I. INTRODUCTION

26 Electromagnons, a coupled oscillation wave of electrical and magnetic dipoles in magneto-
27 electric materials, have bolstered large interest since they were first discussed in 1970s [1, 2].
28 They have remained elusive for a long time, except in a few materials such as rare-earth
29 (R) manganites RMnO₃ [3–5], RMn₂O₅ [6, 7] or ferrite perovskite oxides [8–11]. Among
30 them, BiFeO₃, a room temperature multiferroic [12], has been one of the most intensely
31 considered magnetoelectrics for electromagnon detection, characterization and technological
32 applications. Among the various experimental and theoretical studies of electromagnons,
33 most have focused on so-called electro-active magnons, *i.e.*, control of spin waves by electric
34 fields [13–17]. On the other hand, much less work has been devoted to the study of magnetic
35 control of polarization waves, with a few theoretical works realized in BiFeO₃ [18–20] and
36 manganites [21]. The present work goes one step further by bridging the two approaches, as
37 we intend to demonstrate the possibility of resonantly exciting electromagnons (induced by
38 ferroelectric domain walls) using ac magnetic fields while concurrently manipulating their
39 frequency with dc electric fields.

40 Note that ferroelectric domain walls can now routinely be written, erased and reconfigured
41 using, for instance, PiezoForce Microscopy [22–25]. The ability to generate magnetically
42 polarization waves localized at domain walls, as proposed in Ref. [18], already promises the

43 tantalizing possibility of reconfigurable nanometer size electrical circuits, whose power is
44 switched on and sustained remotely by ac magnetic fields. If now one is able to act on the
45 electrical polarized waves localized at the ferroelectric domain walls, for instance using local
46 electric fields, one could dream of achieving reconfigurable logical elements for computing
47 or detection.

48 In this work, we investigate how dc electric fields affect the domain-wall-induced electro-
49 magnons evidenced in Ref. [18] in multiferroic BiFeO₃. Using Molecular Dynamics based on
50 an *ab-initio* effective Hamiltonian, we reveal that the frequency of these electromagnons is
51 rather sensitive to these dc fields, either increasing or decreasing with them depending on
52 the chosen electromagnon. This latter different behavior (namely, increase *versus* decrease)
53 is found to be correlated with the real-space localization of the optical phonon associated
54 with these electromagnons.

55 II. METHODS

56 Here and as shown in Figure 1, we simulate a multidomain system of BFO with 180°
57 domain configuration inside which two types of domains alternate along the [110] pseudo-
58 cubic direction. The first type of domain, denoted as $D1$, is shown in red in Fig. 1 and
59 possesses electric dipoles aligned along the $[\bar{1}11]$ direction. The second kind of domain,
60 coined $D2$, is displayed in blue in this figure and exhibits electric dipoles lying along the
61 opposite $[1\bar{1}\bar{1}]$ direction. In both domains the magnetic moments and antiferromagnetic
62 (AFM) moments are along the perpendicular $[211]$ and $[0\bar{1}1]$ directions, respectively.

63 The energetics and properties of the studied system are modeled via the use the effective
64 Hamiltonian framework detailed in Ref. [26] within a molecular dynamics (MD) approach
65 [27], in order to obtain dynamical properties. Technically, Newtonian equations of motion
66 are used to investigate the dynamics of the ionic degrees of freedom of this effective Hamil-
67 tonian – that are local modes which are directly proportional to local electric dipoles inside
68 each 5-atom cell; pseudo-vectors representing the antiferrodistortive motions inside such 5-
69 atom cells; and both homogeneous and inhomogeneous strain components. Regarding the
70 dynamics of the magnetic moments, the approach of Ref. [28] is followed, for which such
71 dynamics are treated through the Landau-Lifshitz-Gilbert (LLG) equation [29].

72 DC electric fields with magnitude ranging between 1.0×10^7 V/m and 1.0×10^8 V/m

73 are applied along the $[\bar{1}11]$ direction to such multidomain, that is currently mimicked by
74 using a $24 \times 24 \times 6$ supercell. It should be noted that an electric field of the magnitude of
75 2.0×10^8 V/m is strong enough to reorient all dipole moments along its direction and convert
76 the configuration to monodomain. The MD simulations are conducted at the temperature of
77 10 K. To ensure that the magnetic moments only slightly fluctuate about the same direction
78 during the course of simulations, a dc magnetic field of the magnitude of 245 T is applied
79 along the $[211]$ direction. A lower magnetic field (which is accessible in practical applications)
80 could be applied. In fact, we also performed simulations with a *dc* magnetic field having
81 a magnitude of 2.45 T along the $[211]$ direction, and obtained similar results. Such a large
82 magnetic field along with low temperature were chosen here to avoid large fluctuations of
83 magnetic properties and see the results more clearly. Each MD simulation was performed
84 for 1,000,000 steps of 0.5 fs each, while both homogeneous and inhomogeneous strains were
85 relaxed.

86 III. RESULTS AND DISCUSSION

87 It is important to realize that, under zero dc electric field, there is no macroscopic polar-
88 ization, as a result of the cancellations between the polarizations of the $D1$ and $D2$ domains.
89 On the other hand, we numerically found that the application of our considered dc electric
90 fields results in the increase of the magnitude of electric dipoles in the $D1$ regions and its
91 reduction in the $D2$ areas, therefore yielding now a finite overall polarization along the direc-
92 tion of the applied field which is $[\bar{1}11]$. It is worth mentioning that we observed the change
93 in the structure from a 180-degree multidomain to a monodomain when the applied electric
94 field exceeds a threshold ($E_{threshold} = 2 \times 10^8$ V/m) which is rather abrupt - at least with
95 the resolution of the applied electric field change here, being $E = 1 \times 10^7$ V/m. One may
96 thus need a much smaller step in field's magnitude to observe motion of the domain walls.
97 One can also increase the temperature to see such motion. For instance, and as shown in
98 the Supplementary Material, motion of the domain walls does occur at $T = 300$ K when the
99 applied electric field varies from $E = 3 \times 10^7$ V/m to $E = 5 \times 10^7$ V/m.

100 We then perform Fourier analysis of the temporal evolution of this resulting electrical
101 polarization along the $[\bar{1}11]$ direction, and that of the magnetization along the $[211]$ direction.
102 Figure 2 shows the resulting Fast Fourier Transformations (FFT) of both the electrical

103 polarization and magnetization for a dc electric field of 1.0×10^7 V/m, which reveals the
 104 existence of frequency modes presenting phonons and magnons. Here, in particular, some
 105 peaks are observed at the same frequency in the FFT spectrum of both polarization and
 106 magnetization, which is a signature of electromagnons. Within the electromagnonic modes
 107 that are observed here, four modes are of particular interest due to their noticeable frequency
 108 shift as a response to the application of electric fields of various magnitudes. These modes
 109 are denoted by Mode 1, Mode 2, Mode 3, and Mode 4 in Figure 2 (Note that these four
 110 modes have very similar frequencies than those one can guess to be around 90 cm^{-1} (\simeq
 111 2700 GHz) and 140 cm^{-1} ($\simeq 4200 \text{ GHz}$) in the Supplemental materials of Ref. [30] for
 112 precisely 180° domains of BFO). The electric-field-induced frequency shift of both the phonon
 113 and magnon associated with the electromagnonic Mode 1 is demonstrated in Figs. 3a and
 114 3b, respectively. For this mode, as indicated by purple solid lines in Figs. 3a and 3b,
 115 when the applied electric field is 1.0×10^7 V/m the phonon peak is at 2490 GHz (Figure
 116 3a) and the magnon peak is exactly at the same frequency location (Figure 3b). When
 117 the applied electric field increases to higher magnitudes, the frequency of the phonon and
 118 magnon decrease concurrently which results in a softening for Mode 1. It should be noted
 119 that, in multiferroics, strain can contribute to the emergence of so-called electroacoustic
 120 magnons as a mixture of acoustic phonons, optical phonons, and magnons [19, 20]. Here,
 121 we numerically found (not shown here) that the four aforementioned modes are present in
 122 the frequency spectrum of both the polarization and magnetization even if the homogeneous
 123 strain is clamped during the course of MD simulations, which clarifies that these modes are
 124 electromagnons consisting of optical phonons and magnons. **It should be noted that we**
 125 **do not believe that these computed intensities of Figure 3 have a real physical meaning in**
 126 **our simulations, since such intensities are not monotonic with the fields and can vary when**
 127 **slightly changing some technical details of the Fourier Transform (especially, considering the**
 128 **small values of the vertical scale in the Figure). On the other hand, the frequency position**
 129 **of these peaks is insensitive to such details and does carry physical significance.**

130 For each of these four considered modes, the evolution of their frequency as a function
 131 of the magnitude of the *dc* electric field is shown in Figure 4. One can see that Mode
 132 1 experiences a rather strong decrease of its frequency when the electric field varies from
 133 1.0×10^7 V/m to 1.0×10^8 V/m, namely by about 200 GHz from 2490 GHz to 2290 GHz .
 134 Note that it is known that the effective Hamiltonians of BFO overestimate the magnitude

135 of electric fields by a factor of 23 [31]. Hence, our maximum applied electric field would
 136 correspond experimentally to approximately 43 kV/cm, which is easily sustained in BiFeO₃
 137 thin films [32]. Note also that the decrease of the Mode 1 frequency appears to be quadratic
 138 in nature. Mode 2 also adopts a reduction of its frequency, but at a smaller extent (that
 139 is by about 55 GHz from 2715 GHz to 2660 GHz) and in a linear fashion, with a rate of
 140 0.06 GHz/(kV/cm). Interestingly, such linear variation has indeed been observed in BiFeO₃
 141 for some electromagnons. For instance, the frequency of the so-called extra-cyclon mode ψ_2
 142 may increase or decrease (depending on whether the electric field increases or decreases the
 143 polarization) with a rate of approximately 1.3 GHz/(kV/cm) [33]. In the same reference,
 144 the cyclon mode ϕ_2 shows an opposite behavior with an electrical rate of control of the
 145 magnon frequency of ≈ 0.24 GHz/(kV/cm). Note that we focus here on domain-wall-
 146 induced electromagnons with an antiferromagnetic structure, while the cyclon and extra-
 147 cyclon modes considered in Ref [33] are modes in a monodomain single crystal having a
 148 magnetic cycloidal state. In addition, the commonly known overestimation of electric fields
 149 in effective Hamiltonian models may also contribute to the difference between computational
 150 and experimental rates for the dc-field-induced change in frequency. As a matter of fact,
 151 rescaling our theoretical fields by dividing them by 23 (as indicated in Ref. [31]) results in
 152 a rate for our Mode 2 that goes from 0.06 GHz/(kV/cm) to 1.38 GHz/(kV/cm), which is
 153 similar to the observed magnitude of such rate for the ψ_2 mode in Ref [33]. Similarly, Mode
 154 3 has the same kind of qualitative behavior than Mode 2 but with about twice the slope –
 155 that is, a linear decrease of its frequency by $\simeq 117$ GHz when the *dc* electric field strengthens
 156 from 1.0×10^7 V/m to 1.0×10^8 V/m. Strikingly, Mode 4, whose frequency is basically that
 157 of Mode 3 for an interpolated zero field, adopts a mirror behavior with respect to Mode
 158 3, in the sense that its frequency concomitantly linearly increases by a similar amount of
 159 $\simeq 117$ GHz. Note that we also performed simulations with *opposite dc* electric fields (i.e.,
 160 along $[1\bar{1}\bar{1}]$), which allows us to further determine that the frequencies of Modes 1 and 2
 161 depend on the *magnitude* of the electric field along $[\bar{1}11]$ or $[1\bar{1}\bar{1}]$ while those of Modes 3 and
 162 4 linearly depend on the *projection* of the electric field along $[\bar{1}11]$ (i.e., on the magnitude
 163 but also sign of this projection).

164 In order to understand all these behaviors and demonstrate their relationship with real-
 165 space localization, a layer-by-layer analysis is performed at the different planes that are
 166 parallel to the domain wall. More precisely, the Fourier transform of the average of po-

167 larization of each of these planes is computed for the frequencies associated with the four
168 aforementioned modes for a dc field of 1.0×10^7 V/m, and is shown in Figure 5. For instance,
169 Panel (a) of Figure 5 tells us that Mode 1 is a mode that is strongly localized at the domain
170 walls. Furthermore, Panel (b) of Figure 5 reveals that Mode 2 also localizes near the domain
171 walls but to a smaller extent, as seen by comparing its vertical scale with that of Figure
172 5(a). Consequently, by looking at Figs 4(a), 4(b), 5(a) and 5(b), one can conclude that
173 modes localizing near the domain walls soften under a *dc* electric field, that is they have
174 their frequency decreasing when the field increases – and such decrease is larger when the
175 localization near the walls is stronger.

176 Interestingly, Figures 5(c) and 5(d) tell us that Modes 3 and 4 are rather different from
177 Modes 1 and 2, in the sense that they prefer to localize inside the domains rather than at
178 the domain walls. More precisely, Mode 3 reaches its maximum of the Fourier transform
179 of the polarization in the *D2* region inside which the polarization is antiparallel to the
180 applied electric field. Consequently, applying such field will decrease the magnitude of the
181 polarization in the *D2* area, which corresponds to a local softening of the optical phonon,
182 hence explaining the decrease of the frequency seen in Fig. 4c for Mode 3. In contrast,
183 Mode 4 preferentially localizes in the *D1* area for which the polarization is parallel to the
184 *dc* electric field, and, as a result, such polarization increases in magnitude when the field
185 strengthens. Such increase leads to a local hardening of Mode 4, therefore to a frequency
186 that now increases with the field. Note that Modes 3 and 4 (whose frequency are about
187 4210 GHz and 4245 GHz for a field of 1.0×10^7 V/m, which correspond to 140 cm^{-1} and
188 142 cm^{-1} , respectively) can be thought as both originating from the known zone-center optical
189 phonon of BiFeO₃ monodomain, that then splits in two under dc electric fields because of the
190 existence of domain walls and two different types of domains in our studied system. Based
191 on previous works on BiFeO₃ monodomain single crystals and the fact that we numerically
192 further found (not show here) that Modes 3 and 4 have rather small FFT of the component
193 of the polarization along the [110]-direction (which is perpendicular to the polarizations of
194 both D1 and D2), one can suggest that Modes 3 and 4 originate from the $A_1(LO)$ mode,
195 rather than the $E(TO)$ mode, of BiFeO₃ monodomain [28, 34–37].

196 IV. SUMMARY

197 In summary, we used an atomistic effective Hamiltonian to reveal that the frequency of
198 electromagnons can be significantly tuned by applying dc electric fields in the prototypi-
199 cal BiFeO_3 multiferroic adopting ferroelectric domains. Such finding is promising towards
200 the design of novel devices taking advantage of the dual electric and magnetic natures of
201 electromagnons, with the additional conveniences demonstrated here that (1) it should thus
202 be possible to select the desired operating frequency by choosing the right combination of
203 ac magnetic field's frequency and magnitude of the dc electric field (in order to activate
204 such electromagnons at this desired frequency); and (2) some of these electromagnons are
205 localized at the ferroelectric domain walls, therefore rendering feasible the application of
206 local electric fields for realizing reconfigurable logical elements for computing or detection.
207 These domain-wall-induced electromagnons are further found to either increase or decrease
208 their frequencies under the dc electric fields, depending on the real-space localization of
209 their associated phonons— that is at the ferroelectric domain walls or at the “up” *versus*
210 “down” domains. We therefore hope that the present study deepens the fascinating fields
211 of electromagnons, ferroelectric domains and magnonics.

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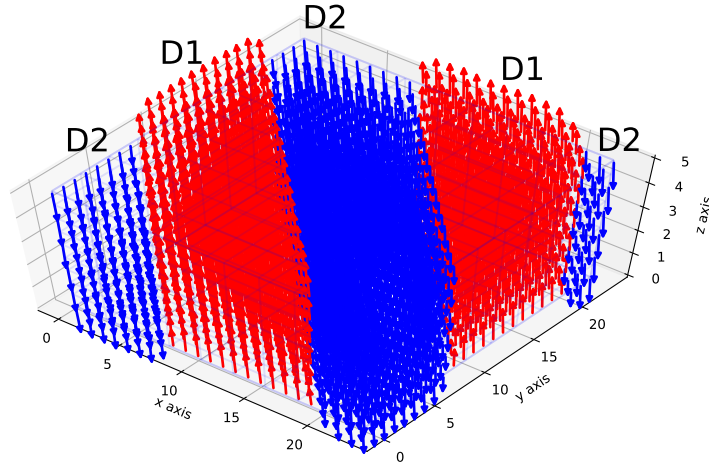


FIG. 1: (Color online) Schematic representation of the zero-field electric dipole moments' pattern for our studied $24 \times 24 \times 6$ supercell of BiFeO_3 . The blue and red vectors are used to represent electric dipole moments along the $[1\bar{1}\bar{1}]$ and $[\bar{1}11]$ directions, respectively.

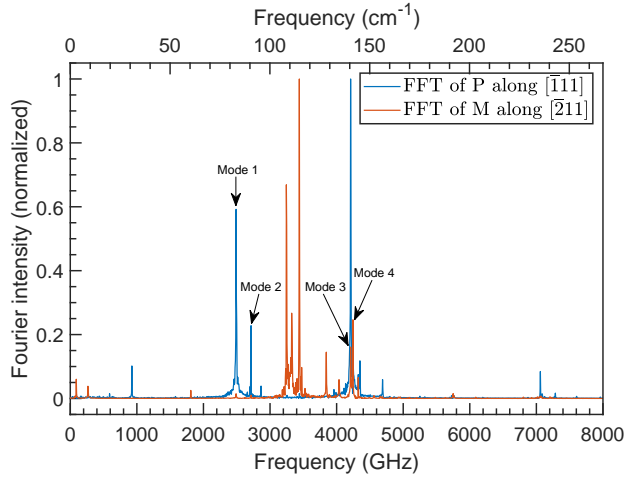


FIG. 2: (Color online) Fourier analyses of polarization and magnetization. The frequency spectrum of the component of electrical dipole moments along the $[\bar{1}11]$ direction (blue) and magnetic moments along the $[211]$ direction (orange) obtained by Fourier analysis when the applied dc electric field is $1 \times 10^7 V/m$. The frequency of the modes shown by black arrows significantly shifts upon applying different magnitudes of electric field.

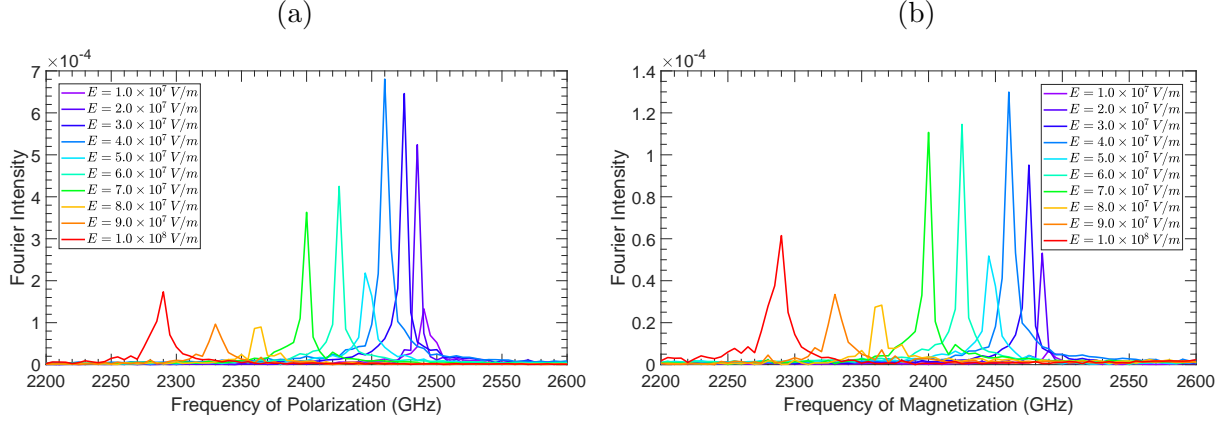


FIG. 3: (Color online) Frequency shift of optical phonons and magnons of Mode 1. (a) The shift of the frequency observed for the optical phonons upon the application of various *dc* electric fields. (b) The shift of the frequency observed for the magnons upon the application of various *dc* electric fields. For both cases the applied electric field changes from 1.0×10^7 V/m to 1.0×10^8 V/m.

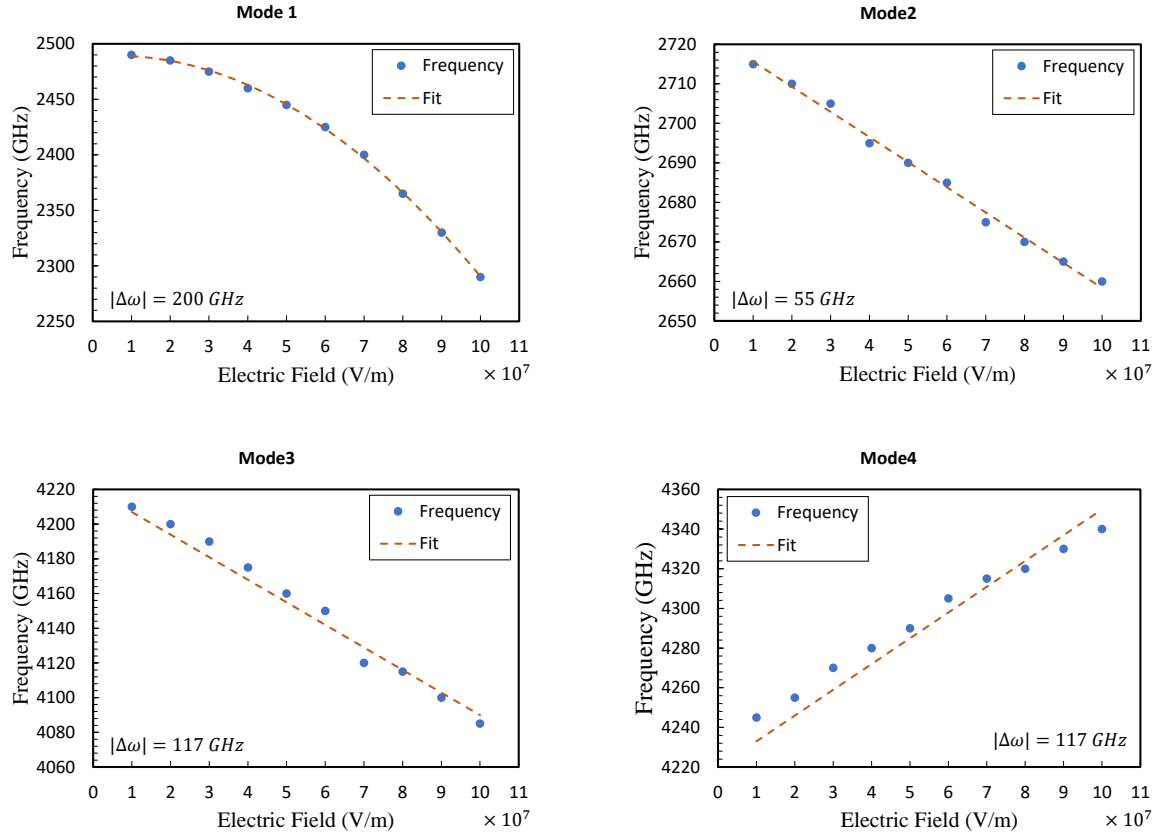


FIG. 4: (Color online) Frequency shift of four electromagnons. In each panel, the frequency shift of the corresponding mode *versus* the applied electric fields is shown. A polynomial fit of second order for Mode 1, and of first order for Modes 2, 3, and 4 is applied. $|\Delta\omega|$ is the magnitude of the difference between the highest and the lowest frequencies of the fitted lines for our chosen range of applied electric fields.

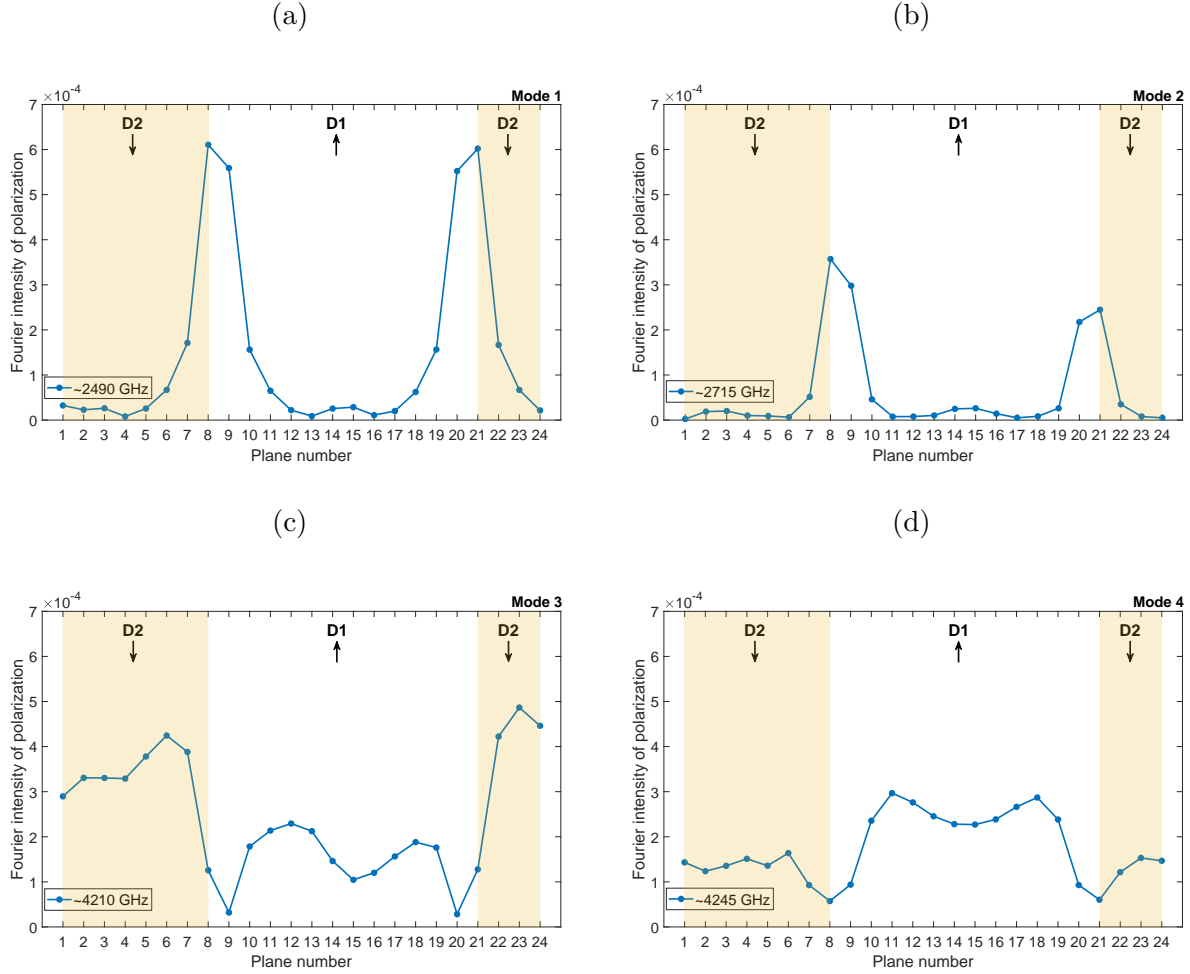


FIG. 5: (Color online) The degree of localization of the electromagnons possessing significant frequency shift under the application of electric fields. In each panel, the degree of localization of the corresponding mode at the planes parallel to the domain walls within the investigated supercell is shown. The shaded area corresponds to the domains possessing electric dipoles pointing along the $[\bar{1}\bar{1}\bar{1}]$ direction, while the white area corresponds to the domains possessing electric dipoles pointing along the opposite $[\bar{1}\bar{1}\bar{1}]$ direction. The arrow in each domain indicates the direction of the z-component of electric dipole moments present in the domain. The applied electric field for all cases is $E = 1.0 \times 10^7 V/m$ along the $[\bar{1}\bar{1}\bar{1}]$ direction which is parallel to the direction of the electric dipoles in D1 and antiparallel to the direction of the electric dipoles in D2.