Supplementary Information of "Strain-induced resonances in the dynamical quadratic magnetoelectric response of multiferroics"

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This Supplementary Material details the derivations of the quadratic magnetoelectric effect discussed in the main article. Also, the contribution of the linear coefficient, α, in the magnetoelectric response is discussed. A demonstration of the validity of the analytical model, via a comparison with the MD simulation results, is provided as well.

Hamiltonian

Let us first consider the following phenomenological Hamiltonian:

$$\mathcal{H} = \frac{1}{2}m_P\dot{P}^2 + \frac{1}{2}m_\eta\dot{\eta}^2 + \frac{1}{2}kP^2 + \frac{1}{4}bP^4 + \frac{1}{2}\epsilon P^2M^2 - EP + \frac{1}{2}Q\eta P^2 + \frac{1}{2}\lambda\eta M^2 + \frac{1}{2}c\eta^2, \quad (1)$$

with P being the polarization, η being the homogeneous strain, M being the magnetization and E being an externally applied electric field.

Dynamical Equations

Using the simplified Hamiltonian in Eq. (1), we can derive the Newtonian equations of motion,

$$m_P\ddot{P} = -kP - bP^3 - \epsilon M^2P + E - Q\eta P - \gamma_P\dot{P}, \quad (2)$$

$$m_\eta\ddot{\eta} = -c\eta - \frac{1}{2}QP^2 - \frac{1}{2}\lambda M^2 - \gamma_\eta\dot{\eta}, \quad (3)$$

in which we included a damping term both for the polarization (γ_P) and the strain (γ_η) Introducing $\omega_P = \sqrt{\frac{k}{m_P}}$, $\omega_\eta = \sqrt{\frac{c}{m_\eta}}$, $\xi_P = \frac{b}{m_P}$, $\xi_{PM} = \frac{\epsilon}{m_P}$, $\Gamma_P = \frac{\gamma_P}{m_P}$, $\tilde{Q}_P = \frac{Q}{m_P}$, $\tilde{Q}_\eta = \frac{Q}{m_\eta}$, $\tilde{\lambda} = \frac{\lambda}{m_\eta}$ and $\Gamma_{eta} = \frac{\gamma_\eta}{m_\eta}$, the equations of motion become

$$\ddot{P} = -\omega_P^2P - \xi_PP^3 - \xi_{PM}M^2P + \frac{1}{m_P}E - \tilde{Q}_P\eta P - \Gamma_P\dot{P} \quad (4)$$

$$\ddot{\eta} = -\omega_\eta^2\eta - \frac{1}{2}\tilde{Q}_\eta P^2 - \frac{1}{2}\tilde{\lambda}M^2 - \Gamma_\eta\dot{\eta} \quad (5)$$

Now, introducing the Fourier transform of the polarization, strain and magnetization:

$$P = \frac{1}{\sqrt{2\pi}} \int d\omega \tilde{P}(\omega) e^{i\omega t}, \tag{6}$$

$$\eta = \frac{1}{\sqrt{2\pi}} \int d\omega \tilde{\eta}(\omega) e^{i\omega t}, \tag{7}$$

$$M = \frac{1}{\sqrt{2\pi}} \int d\omega \tilde{M}(\omega) e^{i\omega t}, \tag{8}$$

make the equations of motion leading to:

$$\begin{aligned}
&\left(\omega_P^2 - \omega^2 + i\Gamma_P\omega\right) \tilde{P} \\
&= -\xi_P \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2) \\
&\quad -\xi_{PM} \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2) \\
&\quad -\tilde{Q}_P \int d\omega_1 \tilde{P}(\omega-\omega_1)\tilde{\eta}(\omega_1) + E
\end{aligned} \tag{9}$$

$$\begin{aligned}
\left(\omega_\eta^2 - \omega^2 + i\Gamma_\eta\omega\right) \tilde{\eta} &= -\frac{1}{2}\tilde{Q}_\eta \int d\omega_1 \tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1) \\
&\quad -\frac{1}{2}\tilde{\lambda} \int d\omega_1 \tilde{M}(\omega-\omega_1)\tilde{M}(\omega_1)
\end{aligned} \tag{10}$$

Now let us insert Eq. (10) into Eq. (9), and assume a vanishing electric field E which results in

$$\begin{aligned}
&\left(\omega_P^2 - \omega^2 + i\Gamma_P\omega\right) \tilde{P} \\
&= -\xi_P \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2) \\
&\quad - \underbrace{\xi_{PM} \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)}_{\text{(Direct ME coupling)}} \\
&\quad + \underbrace{\frac{1}{2}\tilde{Q}_P\tilde{Q}_\eta \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2)}_{\text{(Electrostriction)}} \\
&\quad + \underbrace{\frac{1}{2}\tilde{Q}_P\tilde{\lambda} \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)}_{\text{Electrostriction + Magnetostriction}}
\end{aligned} \tag{11}$$

First derivative

Let us derive Eq. (11) with respect to a magnetic field $H(\omega_0)$ having an angular frequency ω_0. This gives:

$$
\begin{aligned}
&\left(\omega_P^2 - \omega^2 + i\Gamma_P\omega\right)\frac{\partial\tilde{P}}{\partial H(\omega_0)} \\
&= -\xi_P \int d\omega_1 \int d\omega_2 \left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2)\right. \\
&\quad +\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{P}(\omega_2) \\
&\quad \left.+\ \tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2)\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0)}\right] \\
&\quad -\xi_{PM}\int d\omega_1 \int d\omega_2 \left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)\right. \\
&\quad +\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{M}(\omega_2) \\
&\quad \left.+\ \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0)}\right] \\
&\quad +\frac{1}{2}\tilde{Q}_P\tilde{Q}_\eta\int d\omega_1\int d\omega_2\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2)\right. \\
&\quad +\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{P}(\omega_2) \\
&\quad \left.+\ \tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2)\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0)}\right] \\
&\quad +\frac{1}{2}\tilde{Q}_P\tilde{\lambda}\int d\omega_1\int d\omega_2\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)\right. \\
&\quad +\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{M}(\omega_2) \\
&\quad \left.+\ \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0)}\right]
\end{aligned}
\tag{12}
$$

Second derivative

Let us now derive Eq. (12) with respect to a second magnetic field $H(\omega_0')$. We obtain

$$
\begin{aligned}
&\left(\omega_P^2-\omega^2+i\Gamma_P\omega\right)\frac{\partial^2\tilde{P}}{\partial H(\omega_0')\partial H(\omega_0)}\\
&=-\xi_P\int d\omega_1\int d\omega_2\left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')}\tilde{P}(\omega_2)\right.\\
&\quad+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{P}(\omega_1-\omega_2)\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0')}\\
&\quad+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{P}(\omega_2)\\
&\quad+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0)}\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0')}\\
&\quad+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\tilde{P}(\omega_1-\omega_2)\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0)}\\
&\quad+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')}\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0)}\\
&\quad+\frac{\partial^2\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2)\\
&\quad+\tilde{P}(\omega-\omega_1)\frac{\partial^2\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{P}(\omega_2)\\
&\quad\left.+\ \tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2)\frac{\partial^2\tilde{P}(\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\right]\\
&\quad-\xi_{PM}\int d\omega_1\int d\omega_2\left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')}\tilde{M}(\omega_2)\right.\\
&\quad+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{M}(\omega_1-\omega_2)\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0')}\\
&\quad+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{M}(\omega_2)\\
&\quad+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)}\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0')}\\
&\quad+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\tilde{M}(\omega_1-\omega_2)\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0)}\\
&\quad+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')}\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0)}\\
&\quad+\frac{\partial^2\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)\\
&\quad+\tilde{P}(\omega-\omega_1)\frac{\partial^2\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{M}(\omega_2)\\
&\quad\left.+\ \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\frac{\partial^2\tilde{M}(\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\right]
\end{aligned}
$$

$$
\begin{aligned}
&+\frac{1}{2}\tilde{Q}_P\tilde{Q}_\eta\int d\omega_1\int d\omega_2\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')}\tilde{P}(\omega_2)\right.\\
&+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{P}(\omega_1-\omega_2)\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0')}+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{P}(\omega_2)\\
&+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0)}\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0')}+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\tilde{P}(\omega_1-\omega_2)\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0)}\\
&+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')}\frac{\partial\tilde{P}(\omega_2)}{\partial H(\omega_0)}+\frac{\partial^2\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2)\\
&\left.+\tilde{P}(\omega-\omega_1)\frac{\partial^2\tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{P}(\omega_2)+\tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2)\frac{\partial^2\tilde{P}(\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\right]\\
&+\frac{1}{2}\tilde{Q}_P\tilde{\lambda}\int d\omega_1\int d\omega_2\frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\times\left[\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')}\tilde{M}(\omega_2)\right.\\
&+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)}\tilde{M}(\omega_1-\omega_2)\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0')}+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)}\tilde{M}(\omega_2)\\
&+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)}\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0')}+\frac{\partial\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')}\tilde{M}(\omega_1-\omega_2)\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0)}\\
&+\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')}\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0)}+\frac{\partial^2\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2)\\
&\left.+\tilde{P}(\omega-\omega_1)\frac{\partial^2\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{M}(\omega_2)+\tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\frac{\partial^2\tilde{M}(\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\right]
\end{aligned}
\tag{13}
$$

Small field limit

Let us then take the small field limit of Eq. (13), remembering that in that limit,

$$\beta(\omega_0',\omega_0)\delta(\omega-\omega_0-\omega_0')=\frac{1}{2}\frac{\partial^2\tilde{P}(\omega)}{\partial H(\omega_0')\partial H(\omega_0)}, \tag{14}$$

$$\alpha(\omega_0)\delta(\omega-\omega_0)=\frac{\partial\tilde{P}(\omega)}{\partial H(\omega_0)}, \tag{15}$$

$$\chi_M(\omega_0)\delta(\omega-\omega_0)=\frac{\partial\tilde{M}(\omega)}{\partial H(\omega_0)}, \tag{16}$$

$$\chi^{(2)}(\omega_0',\omega_0)\delta(\omega-\omega_0-\omega_0')=\frac{1}{2}\frac{\partial^2\tilde{M}(\omega)}{\partial H(\omega_0')\partial H(\omega_0)}, \tag{17}$$

with $\beta(\omega_0',\omega_0)$ being the quadratic magnetoelectric constant, $\alpha(\omega_0)$ the linear magnetoelectric constant, $\chi_M(\omega_0)$ the magnetic susceptibility and $\chi^{(2)}(\omega_0',\omega_0)$ is the second order magnetic susceptibility.

Let us now calculate each term of Eq. (13). For instance, the first term of Eq. (13) can be rewritten as:

$$\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{\partial \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)} \frac{\partial \tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')} \tilde{P}(\omega_2) \\
&= \int d\omega_1 \int d\omega_2 \alpha(\omega_0)\delta(\omega-\omega_1-\omega_0)\alpha(\omega_0')\delta(\omega_1-\omega_2-\omega_0')\tilde{P}(\omega_2) \\
&= \int d\omega_2 \alpha(\omega_0)\alpha(\omega_0')\delta(\omega-\omega_0-\omega_0'-\omega_2)\tilde{P}(\omega_2) \\
&= \alpha(\omega_0)\alpha(\omega_0')\tilde{P}(\omega-\omega_0-\omega_0')
\end{aligned} \tag{18}$$

We also obtain:

$$\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{\partial^2 \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')\partial H(\omega_0)} \tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2) \\
&= \int d\omega_1 \int d\omega_2 2\beta(\omega_0',\omega_0)\delta(\omega-\omega_1-\omega_0-\omega_0')\tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2) \\
&= 2\beta(\omega_0',\omega_0) \int d\omega_2 \tilde{P}(\omega-\omega_0-\omega_0'-\omega_2)\tilde{P}(\omega_2).
\end{aligned} \tag{19}$$

$$\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{\partial \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)} \frac{\partial \tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')} \tilde{M}(\omega_2) \\
&= \int d\omega_1 \int d\omega_2 \alpha(\omega_0)\delta(\omega-\omega_1-\omega_0)\chi_M(\omega_0')\delta(\omega_1-\omega_2-\omega_0')\tilde{M}(\omega_2) \\
&= \alpha(\omega_0)\chi_M(\omega_0')\tilde{M}(\omega-\omega_0-\omega_0')
\end{aligned} \tag{20}$$

$$\int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1) \frac{\partial \tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)} \frac{\partial \tilde{M}(\omega_2)}{\partial H(\omega_0')}$$

$$= \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\chi_M(\omega_0)\delta(\omega_1-\omega_2-\omega_0)\chi_M(\omega_0')\delta(\omega_2-\omega_0') \tag{21}$$

$$= \tilde{P}(\omega-\omega_0-\omega_0')\chi_M(\omega_0)\chi_M(\omega_0') \tag{22}$$

$$\begin{aligned}
&\int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2) \frac{\partial^2 \tilde{M}(\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)} \\
&= \int d\omega_1 \int d\omega_2 \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)2\chi^{(2)}(\omega_0',\omega_0)\delta(\omega_2-\omega_0-\omega_0') \\
&= 2\chi^{(2)}(\omega_0',\omega_0) \int d\omega_1 \tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_0-\omega_0')
\end{aligned} \tag{23}$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{\partial^2 \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0')\partial H(\omega_0)} \tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2) \\
&= \int d\omega_1 \int d\omega_2 2\beta(\omega_0',\omega_0)\delta(\omega-\omega_1-\omega_0-\omega_0')\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2) \\
&= 2\beta(\omega_0',\omega_0)\int d\omega_2 \tilde{M}(\omega-\omega_0-\omega_0'-\omega_2)\tilde{M}(\omega_2)
\end{aligned}
\tag{24}
$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \frac{\partial \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)} \frac{\partial \tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')} \tilde{P}(\omega_2) \\
&= \int d\omega_1 \int d\omega_2 \frac{\alpha(\omega_0)\delta(\omega-\omega_1-\omega_0)\alpha(\omega_0')\delta(\omega_1-\omega_2-\omega_0')\tilde{P}(\omega_2)}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \\
&= \int d\omega_2 \frac{\alpha(\omega_0)\alpha(\omega_0')\tilde{P}(\omega_2)\delta(\omega-\omega_0-\omega_0'-\omega_2)}{\omega_\eta^2-(\omega-\omega_0)^2+i\Gamma_\eta(\omega-\omega_0)} \\
&= \frac{\alpha(\omega_0)\alpha(\omega_0')\tilde{P}(\omega-\omega_0-\omega_0')}{\omega_\eta^2-(\omega-\omega_0)^2+i\Gamma_\eta(\omega-\omega_0)}
\end{aligned}
\tag{25}
$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \frac{\partial \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)} \tilde{P}(\omega_1-\omega_2) \frac{\partial \tilde{P}(\omega_2)}{\partial H(\omega_0')} \\
&= \int d\omega_2 \frac{\alpha(\omega_0)\alpha(\omega_0')\tilde{P}(\omega_2)\delta(\omega-\omega_0-\omega_0'-\omega_2)}{\omega_\eta^2-(\omega-\omega_0)^2+i\Gamma_\eta(\omega-\omega_0)} \\
&= \frac{\alpha(\omega_0)\alpha(\omega_0')\tilde{P}(\omega-\omega_0-\omega_0')}{\omega_\eta^2-(\omega-\omega_0)^2+i\Gamma_\eta(\omega-\omega_0)}
\end{aligned}
\tag{26}
$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \tilde{P}(\omega-\omega_1) \frac{\partial \tilde{P}(\omega_2)}{\partial H(\omega_0)} \frac{\partial \tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0')} \\
&= \int d\omega_1 \int d\omega_2 \frac{\tilde{P}(\omega-\omega_1)\alpha(\omega_0)\delta(\omega_1-\omega_2-\omega_0)\alpha(\omega_0')\delta(\omega_2-\omega_0')}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \\
&= \int d\omega_1 \frac{\tilde{P}(\omega-\omega_1)\alpha(\omega_0)\alpha(\omega_0')\delta(\omega_0+\omega_0'-\omega_1)}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \\
&= \frac{\alpha(\omega_0)\alpha(\omega_0')\tilde{P}(\omega-\omega_0-\omega_0')}{\omega_\eta^2-(\omega_0+\omega_0')^2+i\Gamma_\eta(\omega_0+\omega_0')}
\end{aligned}
\tag{27}
$$

$$\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1} \frac{\partial^2 \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)\partial H(\omega_0')} \tilde{P}(\omega_1-\omega_2)\tilde{P}(\omega_2)$$
$$= \int d\omega_1 \int d\omega_2 \frac{2\beta(\omega_0,\omega_0')\tilde{P}(\omega_1-\omega_2)\tilde{\omega_2}\delta(\omega-\omega_1-\omega_0-\omega_0')}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1}$$
$$= \frac{2\beta(\omega_0,\omega_0')}{\omega_\eta^2 - (\omega-\omega_0-\omega_0')^2 + i\Gamma_\eta(\omega-\omega_0-\omega_0')} \int d\omega_2 \tilde{P}(\omega-\omega_0-\omega_0'-\omega_2)\tilde{P}(\omega_2) \quad (28)$$

$$\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1} \tilde{P}(\omega-\omega_1) \frac{\partial^2 \tilde{P}(\omega_1-\omega_2)}{\partial H(\omega_0)\partial H(\omega_0')} \tilde{P}(\omega_2)$$
$$= \int d\omega_1 \int d\omega_2 \frac{2\beta(\omega_0,\omega_0')\delta(\omega_1-\omega_2-\omega_0-\omega_0')\tilde{P}(\omega-\omega_1)\tilde{P}(\omega_2)}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1}$$
$$= 2\beta(\omega_0,\omega_0') \int d\omega_1 \frac{\tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_0-\omega_0')}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1} \quad (29)$$

Similarly,

$$\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1} \tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_2) \frac{\partial^2 \tilde{P}(\omega_2)}{\partial H(\omega_0)\partial H(\omega_0')}$$
$$= 2\beta(\omega_0,\omega_0') \int d\omega_1 \frac{\tilde{P}(\omega-\omega_1)\tilde{P}(\omega_1-\omega_0-\omega_0')}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1} \quad (30)$$

$$\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1} \frac{\partial \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)} \frac{\partial \tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')} \tilde{M}(\omega_2)$$
$$= \int d\omega_1 \int d\omega_2 \frac{\alpha(\omega_0)\delta(\omega-\omega_1-\omega_0)\chi_M(\omega_0')\delta(\omega_1-\omega_2-\omega_0')\tilde{M}(\omega_2)}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1}$$
$$= \int d\omega_2 \frac{\alpha(\omega_0)\chi_M(\omega_0')\tilde{M}(\omega_2)\delta(\omega-\omega_0-\omega_0'-\omega_2)}{\omega_\eta^2 - (\omega-\omega_0)^2 + i\Gamma_\eta(\omega-\omega_0)} \quad (31)$$
$$= \frac{\alpha(\omega_0)\chi_M(\omega_0')\tilde{M}(\omega-\omega_0-\omega_0')}{\omega_\eta^2 - (\omega-\omega_0)^2 + i\Gamma_\eta(\omega-\omega_0)} \quad (32)$$

$$\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta \omega_1} \frac{\partial \tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)} \tilde{M}(\omega_1-\omega_2) \frac{\partial \tilde{M}(\omega_2)}{\partial H(\omega_0')}$$
$$= \frac{\alpha(\omega_0)\chi_M(\omega_0')\tilde{M}(\omega-\omega_0-\omega_0')}{\omega_\eta^2 - (\omega-\omega_0)^2 + i\Gamma_\eta(\omega-\omega_0)} \quad (33)$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\tilde{P}(\omega-\omega_1)\frac{\partial\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0)}\frac{\partial\tilde{M}(\omega_2)}{\partial H(\omega_0')} \\
&=\int d\omega_1 \int d\omega_2 \frac{\tilde{P}(\omega-\omega_1)\chi_M(\omega_0)\delta(\omega_1-\omega_2-\omega_0)\chi_M(\omega_0')\delta(\omega_2-\omega_0')}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \\
&=\frac{\tilde{P}(\omega-\omega_0-\omega_0')\chi_M(\omega_0)\chi_M(\omega_0')}{\omega_\eta^2-(\omega_0+\omega_0')^2+i\Gamma_\eta(\omega_0+\omega_0')}
\end{aligned}
\tag{34}
$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\frac{\partial^2\tilde{P}(\omega-\omega_1)}{\partial H(\omega_0)\partial H(\omega_0')}\tilde{M}(\omega_1-\omega_2)\tilde{M}(\omega_2) \\
&=\int d\omega_1 \int d\omega_2 \frac{2\beta(\omega_0,\omega_0')\delta(\omega-\omega_1-\omega_0-\omega_0')}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \\
&=\frac{2\beta(\omega_0,\omega_0')}{\omega_\eta^2-(\omega-\omega_0-\omega_0')^2+i\Gamma_\eta(\omega-\omega_0-\omega_0')}
\end{aligned}
\tag{35}
$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\tilde{P}(\omega-\omega_1)\frac{\partial^2\tilde{M}(\omega_1-\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)}\tilde{M}(\omega_2) \\
&=\int d\omega_1 \int d\omega_2 \frac{\tilde{P}(\omega-\omega_1)2\chi_M^{(2)}(\omega_0',\omega_0)\delta(\omega_1-\omega_2-\omega_0-\omega_0')\tilde{M}(\omega_2)}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \\
&=2\chi_M^{(2)}(\omega_0',\omega_0)\int d\omega_1 \frac{\tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_0-\omega_0')}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}
\end{aligned}
\tag{36}
$$

$$
\begin{aligned}
&\int d\omega_1 \int d\omega_2 \frac{1}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)\frac{\partial^2\tilde{M}(\omega_2)}{\partial H(\omega_0')\partial H(\omega_0)} \\
&=\int d\omega_1 \int d\omega_2 \frac{\tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_2)2\chi_M^{(2)}(\omega_0',\omega_0)\delta(\omega_2-\omega_0-\omega_0')}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1} \\
&=2\chi_M^{(2)}(\omega_0',\omega_0)\int d\omega_1 \frac{\tilde{P}(\omega-\omega_1)\tilde{M}(\omega_1-\omega_0-\omega_0')}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}
\end{aligned}
\tag{37}
$$

As a result, from Eq. (13), one gets the following equation satisfied by β, and further requiring that $\omega_0'+\omega_0=\omega$ (recalling also that P, M are real quantities, which therefore implies that $\tilde{P}(-\omega)=\tilde{P}^\star(\omega)$):

$$
\begin{aligned}
&\left(\omega_P^2-(\omega_0+\omega_0')^2+i\Gamma_P(\omega_0+\omega_0')\right)2\beta(\omega_0',\omega_0)\\
&=-2\xi_P\left[3\alpha(\omega_0')\alpha(\omega_0)\tilde{P}(0)+3\beta(\omega_0',\omega_0)\int d\omega_2|\tilde{P}(\omega_2)|\right]\\
&\quad-2\xi_{PM}\left[\left(\alpha(\omega_0)\chi_M(\omega_0')+\alpha(\omega_0')\chi_M(\omega_0)\right)\tilde{M}(0)+\chi_M(\omega_0')\chi_M(\omega_0)\tilde{P}(0)\right.\\
&\quad\left.\beta(\omega_0',\omega_0)\int d\omega_2|\tilde{M}(\omega_2)|+2\chi_M^{(2)}(\omega_0',\omega_0)\int d\omega_1\tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)\right]\\
&\quad+\tilde{Q}_P\tilde{Q}_\eta\left[\alpha(\omega_0')\alpha(\omega_0)\tilde{P}(0)\left(\frac{1}{\omega_\eta^2-\omega_0'^2+i\Gamma_\eta\omega_0'}+\frac{1}{\omega_\eta^2-\omega_0^2+i\Gamma_\eta\omega_0}+\right.\right.\\
&\quad\left.+\frac{1}{\omega_\eta^2-(\omega_0'+\omega_0))^2+i\Gamma_\eta(\omega_0'+\omega_0)}\right)\\
&\quad\left.+\frac{\beta(\omega_0',\omega_0)}{\omega_\eta^2}\int d\omega_2|\tilde{P}(\omega_2)|^2+2\beta(\omega_0',\omega_0)\int d\omega_1\frac{|\tilde{P}(\omega-\omega_1)|^2}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\right]\\
&\quad+\tilde{Q}_P\tilde{\lambda}\left[\frac{\alpha(\omega_0)\chi_M(\omega_0')\tilde{M}(0)}{\omega_\eta^2-\omega_0'^2+i\Gamma_\eta\omega_0'}+\frac{\alpha(\omega_0')\chi_M(\omega_0)\tilde{M}(0)}{\omega_\eta^2-\omega_0^2+i\Gamma_\eta\omega_0}\right.\\
&\quad+\frac{\chi_M(\omega_0)\chi_M(\omega_0')\tilde{P}(0)}{\omega_\eta^2-(\omega_0'+\omega_0)^2+i\Gamma_\eta(\omega_0'+\omega_0)}\\
&\quad\left.\frac{\beta(\omega_0',\omega_0)}{\omega_\eta^2}\int d\omega_2|\tilde{M}(\omega_2)|^2+2\chi_M^{(2)}(\omega_0',\omega_0)\int d\omega_1\frac{\tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\right].
\end{aligned}
\tag{38}
$$

Once all the terms in β have been gathered, we obtain, after introducing

$$
\begin{aligned}
&\tilde{\omega}_P^2\\
&=\omega_P^2+\left(3\xi_P-\frac{\tilde{Q}_P\tilde{\lambda}}{2\omega_\eta^2}\right)\int d\omega_2\left|\tilde{P}(\omega_2)\right|^2+\left(\xi_{PM}-\frac{\tilde{Q}_P\tilde{Q}_\eta}{2\omega_\eta^2}\right)\int d\omega_2\left|\tilde{M}(\omega_2)\right|^2\\
&-\tilde{Q}_P\tilde{Q}_\eta\int d\omega_1\frac{\tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}
\end{aligned}
\tag{39}
$$

$$
\begin{aligned}
&\beta(\omega_0', \omega_0) \\
&= \frac{1}{\tilde{\omega}_P^2 - \omega^2 + i\Gamma_P\omega} \times \\
&\Big[-3\xi_P \alpha(\omega_0')\alpha(\omega_0)\tilde{P}(0) - \xi_{PM}\Big((\alpha(\omega_0)\chi_M(\omega_0') + \alpha(\omega_0')\chi_M(\omega_0))\tilde{M}(0) + \chi_M(\omega_0')\chi_M(\omega_0)\tilde{P}(0) \\
&+2\chi_M^{(2)}(\omega_0', \omega_0)\int d\omega_1 \tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)\Big) \\
&\frac{\tilde{Q}_P\tilde{Q}_\eta}{2}\alpha(\omega_0')\alpha(\omega_0)\tilde{P}(0) \times \left(\frac{1}{\omega_\eta^2 - \omega_0'^2 + i\Gamma_\eta\omega_0'} + \frac{1}{\omega_\eta^2 - \omega_0^2 + i\Gamma_\eta\omega_0} + \frac{1}{\omega_\eta^2 - (\omega_0' + \omega_0))^2 + i\Gamma_\eta(\omega_0' + \omega_0)}\right) \\
&\frac{\tilde{Q}_P\tilde{\lambda}}{2}\Bigg(\frac{\alpha(\omega_0)\chi_M(\omega_0')\tilde{M}(0)}{\omega_\eta^2 - \omega_0'^2 + i\Gamma_\eta\omega_0'} + \frac{\alpha(\omega_0')\chi_M(\omega_0)\tilde{M}(0)}{\omega_\eta^2 - \omega_0^2 + i\Gamma_\eta\omega_0} + \frac{\chi_M(\omega_0)\chi_M(\omega_0')\tilde{P}(0)}{\omega_\eta^2 - (\omega_0' + \omega_0)^2 + i\Gamma_\eta(\omega_0' + \omega_0)} \\
&+2\chi_M^{(2)}(\omega_0', \omega_0)\int d\omega_1 \frac{\tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)}{\omega_\eta^2 - \omega_1^2 + i\Gamma_\eta\omega_1}\Bigg)\Big]
\end{aligned} \tag{40}
$$

Now, in order to simplify a little bit the picture, let us neglect the term proportional or quadratic in the linear ME coefficient α (which will demonstrate to be negligible for large magnetic fields in BFO) and the second order magnetic susceptibility $\chi^{(2)}$. This allows to clearly see the physics and resonances.

As a matter of fact, under those simplifications, the latest equation for β can be re-written as:

$$
\beta(\omega_0', \omega_0) = -\xi_{PM}\frac{\chi_M(\omega_0')\chi_M(\omega_0)\tilde{P}(0)}{\tilde{\omega}_P^2 - \omega^2 + i\Gamma_P\omega} + \frac{\tilde{Q}_P\tilde{\lambda}}{2}\frac{\chi_M(\omega_0')\chi_M(\omega_0)\tilde{P}(0)}{(\tilde{\omega}_P^2 - \omega^2 + i\Gamma_P\omega)(\omega_\eta^2 - (\omega_0' + \omega_0)^2 + i\Gamma_\eta(\omega_0' + \omega_0))} \tag{41}
$$

Now, in the specific case where we applied both a *DC* (hence 0 frequency) and an *ac* field (with frequency ω), we have two components of β to consider.

First of all,

$$
\beta(0, \omega) = -\xi_{PM}\frac{\chi_M(0)\chi_M(\omega)\tilde{P}(0)}{\tilde{\omega}_P^2 - \omega^2 + i\Gamma_P\omega} + \frac{\tilde{Q}_P\tilde{\lambda}}{2}\frac{\chi_M(0)\chi_M(\omega)\tilde{P}(0)}{(\tilde{\omega}_P^2 - \omega^2 + i\Gamma_P\omega)(\omega_\eta^2 - \omega^2 + i\Gamma_\eta\omega)} \tag{42}
$$

is the quadratic ME coupling that couples the DC and the *ac* field to produce a frequency at ω (*i.e.*, is an effective linear ME term). One observes from the equation above that resonances can occur:

1. If $\chi_M(\omega)$ itself has a resonance (which we do not observe in the clamped case in our numerical simulations of $\beta(0, \omega)$ in Figure 2a of the main text).

2. If the *ac* field frequency matches the modified phonon frequency, *i.e.* $\omega = \tilde{\omega}_P$.

3. If the strain resonance is achieved, *i.e.* $\omega = \omega_\eta$, which should only be observed in the unclamped case, as confirmed in Figure 2a in the main text.

Secondly, there is another component of β which couples the *ac* field with itself and produces a doubling in frequency 2ω,

$$\beta(\omega,\omega) = -\xi_{PM}\frac{\chi_M(\omega)^2\tilde{P}(0)}{\tilde{\omega}_P^2 - 4\omega^2 + i\Gamma_P 2\omega} + \frac{\tilde{Q}_P\tilde{\lambda}}{2}\frac{\chi_M(\omega)^2\tilde{P}(0)}{(\tilde{\omega}_P^2 - 4\omega^2 + i\Gamma_P 2\omega)(\omega_\eta^2 - 4\omega^2 + i\Gamma_\eta 2\omega)} \quad (43)$$

Now, we see that there are again three possible resonances:

1. If $\chi_M(\omega)$ has a resonance, $\beta(0,\omega)$ and $\beta(\omega,\omega)$ should have a resonance *at the same frequency* of the applied *ac* field, *i.e.* $\omega \approx \omega_{magnon}$.

2. The resonance frequency in $\beta(\omega,\omega)$ due to optical phonons happens at half the phonon frequency (for $\omega = \frac{\tilde{\omega}_P}{2}$), contrary to $\beta(0,\omega)$, for which it happens at $\tilde{\omega}_P$.

3. There is also a strain resonance in $\beta(\omega,\omega)$. Just like the phonon case above, it should happen at half the strain proper frequency, *i.e.* for $\omega = \frac{\tilde{\omega}_\eta}{2}$; on the other hand, that resonance occurs in $\beta(0,\omega)$ when the frequency of the applied field is $\omega = \omega_\eta$.

Linear ME constant

In this section, we also derive an expression for the linear magnetoelectric coefficient, $\alpha(\omega)$. Taking the small field limit of Eq. (12), we have

$$\begin{aligned}\left(\omega_P^2 - \omega^2 + i\Gamma_P\omega\right)\alpha(\omega) = &-3\xi_P\alpha(\omega)\int d\omega_2\left|\tilde{P}(\omega_2)\right|^2 \\ &-\xi_{PM}\left[\alpha(\omega)\int d\omega_2\left|\tilde{M}(\omega_2)\right|^2 + 2\chi_M(\omega)\int d\omega_1\tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)\right] \\ &+\frac{\tilde{Q}_P\tilde{Q}_\eta}{2}\left[\frac{\alpha(\omega)}{\omega_\eta^2}\int d\omega_2\left|\tilde{P}(\omega_2)\right|^2 + 2\alpha(\omega)\int d\omega_1\frac{\left|\tilde{P}(\omega-\omega_1)\right|^2}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\right] \\ &+\frac{\tilde{Q}_P\tilde{\lambda}}{2}\left[\frac{\alpha(\omega)}{\omega_\eta^2}\int d\omega_2\left|\tilde{M}(\omega_2)\right|^2 + 2\chi_M(\omega)\int d\omega_1\frac{\tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}\right]\end{aligned} \quad (44)$$

Introducing $\tilde{\omega}_P$ as in the previous section, we thus find the following expression for the dynamical linear magnetoelectric constant:

$$\alpha(\omega) = \frac{-2\xi_{PM}\chi_M(\omega)\int d\omega_1 \tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1) + \tilde{Q}_P\tilde{\lambda}\chi_M(\omega)\int d\omega_1 \frac{\tilde{P}(\omega-\omega_1)\tilde{M}^\star(\omega-\omega_1)}{\omega_\eta^2-\omega_1^2+i\Gamma_\eta\omega_1}}{\tilde{\omega_P}^2-\omega^2+i\Gamma_P\omega}\chi_M(\omega) \tag{45}$$

Let us consider a limiting case, for which, prior to excitation, the spectra of polarization and magnetization reduce to a spontaneous polarization P_0 and magnetization M_0; in other words, $P(\omega) = P_0\delta(\omega)$ and $M(\omega) = M_0\delta(\omega)$. In such case, the linear magnetoelectric coefficient reduces to

$$\alpha(\omega) = -2\xi_{PM}\chi_M(\omega)\frac{P_0M_0}{\tilde{\omega}_P^2-\omega^2+i\Gamma_P\omega} + \tilde{Q}_P\tilde{\lambda}\chi_M(\omega)\frac{P_0M_0}{\left(\tilde{\omega_P}^2-\omega^2+i\Gamma_P\omega\right)\left(\tilde{\omega}_\eta^2-\omega^2+i\Gamma_\eta\omega\right)} \tag{46}$$

It can be observed that the first term is an electromagnon-like resonance term, as it depends only on the direct coupling of polarization and magnetization, and can exhibit resonances at the magnon and polar optical phonon frequencies. We note that it has a similar form to the electromagnetic susceptibility derived in Equation 28 of Ref. [1], albeit with a renormalized optical phonon frequencies and the magnetic excitation frequencies hidden in the magnetic susceptibility $\chi_M(\omega)$ in our case. The second term is the product of electrostrictive and magnetostrictive couplings, and shows resonance at the same frequencies as the first term, plus an extra-resonance at the strain frequency. It thus represents the effect of an electroacoustic magnon.

There is a body of work on strain-mediated enhanced magnetoelectric coupling that shows the appearance of specific resonances in the magnetoelectric coupling due to acoustic deformation [2, 3]. It has also been discussed that the combined effect of piezoelectric and pseudopiezomagnetic effects lead to large linear magnetoelectric coefficient [4, 5], or electric-field control of electromagnon frequencies [6]. All of those works focus on ferroelectric-ferromagnetic heterostructures, hence the results are heavily dependent on the quality of the interface between the two films [7]. In contrast, we consider a single phase material, for which the strain resonance can be tuned by changing the size and shape of the material, whilst still leading to strain-mediated resonance. This represents potentially easier processing, and cheaper costs.

The investigation of the contribution of the linear coefficient, α, in the magnetoelectric response

In order to quantify the contribution of the linear coefficient, α, in the magnetoelectric response of $BiFeO_3$, we derive both linear and quadratic coefficients as described in the following:

$$P = P^s + \alpha H + \frac{1}{2}\beta H^2 \tag{47}$$

Here P is electrical polarization, P^s is the spontaneous polarization, and H is the external applied magnetic field. α and β are the linear and quadratic magnetoelectric coefficients, respectively. If the applied magnetic field is a combination of a *dc* field and an *ac* field with the frequency of ω

$$H = H_{dc} + h_{ac}e^{i\omega t} \tag{48}$$

then

$$P = [P^s + \alpha_0 H_{dc} + \frac{1}{2}\beta_0 H_{dc}^2] + [\alpha(\omega)h_{ac} + \beta(0,\omega)H_{dc}h_{ac}]e^{i\omega t} + \frac{1}{2}\beta(\omega,\omega)h_{ac}^2 e^{2i\omega t}. \tag{49}$$

By introducing

$$P_0 = P^s + \alpha_0 H_{dc} + \frac{1}{2}\beta_0 H_{dc}^2 \tag{50}$$

we get

$$P = P_0 + [\alpha(\omega)h_{ac} + \beta(0,\omega)H_{dc}h_{ac}]e^{i\omega t} + [\frac{1}{2}\beta(\omega,\omega)h_{ac}^2]e^{2i\omega t}. \tag{51}$$

Arranging Eq. (51) in the form of $P = P_0 + ae^{i(\omega t+\varphi_\omega)} + ae^{i(2\omega t+\varphi_{2\omega})}$, gives

$$[\alpha'(\omega) + i\alpha''(\omega)]h_{ac} + [\beta^{'}(0,\omega) + i\beta^{''}(0,\omega)]H_{dc}h_{ac} = a\cos\varphi_\omega + i\ a\ sin\ \varphi_\omega \tag{52}$$

$$(\beta^{'}(\omega,\omega) + i\beta^{''}(\omega,\omega))[\frac{1}{2}h_{ac}^2] = b\cos\varphi_{2\omega} + i\ b\sin\varphi_{2\omega} \tag{53}$$

Solving Eq. (52) and Eq. (53) results in

$$\beta^{'}(0,\omega) = \frac{a\cos\varphi_\omega - \alpha^{'}(\omega)h_{ac}}{H_{dc}h_{ac}} = \frac{[\frac{2}{L}\int_0^L (P-P_0)sin(\omega t)dt] - \alpha^{'}(\omega)h_{ac}}{H_{dc}h_{ac}} \tag{54}$$

$$\beta^{''}(0,\omega) = \frac{a\ sin\ \varphi_\omega - \alpha^{''}(\omega)h_{ac}}{H_{dc}h_{ac}} = \frac{[\frac{2}{L}\int_0^L (P-P_0)\cos(\omega t)dt] - \alpha^{''}(\omega)h_{ac}}{H_{dc}h_{ac}} \tag{55}$$

$$\beta^{'}(\omega,\omega) = \frac{b cos\varphi_{2\omega}}{\frac{1}{2}h_{ac}^2} = \frac{P_{2\omega}^{'}}{\frac{1}{2}h_{ac}^2} = \frac{\frac{2}{L}\int_0^L (P-P_0)cos(2\omega t)dt}{\frac{1}{2}h_{ac}^2} \tag{56}$$

$$\beta^{''}(\omega,\omega) = \frac{b sin\varphi_{2\omega}}{\frac{1}{2}h_{ac}^2} = \frac{P_{2\omega}^{''}}{\frac{1}{2}h_{ac}^2} = \frac{\frac{2}{L}\int_0^L (P-P_0)sin(2\omega t)dt}{\frac{1}{2}h_{ac}^2} \tag{57}$$

If we introduce

$$S = \frac{2}{L}\int_0^L (P-P_0)sin(\omega t)dt \tag{58}$$

Then we can rearrange Eq. (54) into the following form

$$H_{dc}h_{ac}\beta^{'}(0,\omega) = S - \alpha^{'}(\omega)h_{ac} \tag{59}$$

$$S = H_{dc}h_{ac}\beta^{'}(0,\omega) + \alpha^{'}(\omega)h_{ac} \tag{60}$$

$$\frac{S}{h_{ac}} = \beta^{'}(0,\omega)H_{dc} + \alpha^{'}(\omega) \tag{61}$$

Now, if we calculate, from our MD simulations, $\frac{S}{h_{ac}}$ for different values of H_{dc} (for a fixed ω and a fixed h_{ac}) and apply a fit based on Equation Eq. (61), we can extract $\alpha^{'}(\omega)$ in addition to $\beta^{'}(0,\omega)$. The results for $\frac{S}{h_{ac}}$ for different values of dc magnetic fields when $h_{ac} = 61.2\,T$ and $\omega = 160\,GHz$ are shown in Figure 1. The goodness of a straight line fit confirms the validity of Eq. (61), and the extracted values are:
$\beta'(0,\omega) = -2.614 \times 10^{-8}$
$\alpha'(\omega) = -3.655 \times 10^{-7}$
The value of $\beta'(0,\omega)H_{dc}$ is thus larger than $\alpha'(\omega)$ by one order of magnitude when H_{dc} is of the order of $100\,T$ or greater. In fact, When the applied magnetic field increases, the contribution of the linear ME response, α, becomes negligible in comparison with the quadratic response such a way that $\beta H_{dc} >> \alpha$.

Numerical demonstration of the validity of the analytical model

In order to demonstrate the validity of our analytical model described by Eq. (42) and Eq. (43), we used it to fit the results of our MD simulations when the frequency of applied magnetic field changes from $20\,GHz$ to $500\,GHz$.

Based on the existence of two electroacoustic magnons, the function defined for $\beta(0,\omega)$ based on Equation Eq. (42) can be modified as follows

$$f(0,\omega) = \frac{A}{\omega_P^2 - \omega^2 + i\Gamma_P\omega} + \frac{B_1}{(\omega_P^2 - \omega^2 + i\Gamma_P\omega)(\omega_{\eta_1}^2 - \omega^2 + i\Gamma_{\eta_1}\omega)} + \frac{B_2}{(\omega_P^2 - \omega^2 + i\Gamma_P\omega)(\omega_{\eta_2}^2 - \omega^2 + i\Gamma_{\eta_2}\omega)} \tag{62}$$

Here A, B_1, B_2, Γ_P , Γ_{η_1}, Γ_{η_2}, ω_{η_1}, and ω_{η_2} are fitting parameters, while ω_P is the frequency of phonon which is set to $4320\, GHz$. Figure 2a of this Supplementary Information clearly demonstrates that the function $f(0,\omega)$ fits very well the MD data for $\beta'(0,\omega)$– therefore confirming the validity of the model leading to Eq. (42).

For the case of $\beta(\omega,\omega)$ a similar approach (based on the existence of two electroacoustic magnons and starting from Eq. (43)) can be used, which results in:

$$f(\omega,\omega) = \frac{A'}{\omega_P^2 - 4\omega^2 + i\Gamma_P 2\omega} + \frac{B_1'}{(\omega_P^2 - 4\omega^2 + i\Gamma_P 2\omega)(\omega_{\eta_1}^2 - 4\omega^2 + i\Gamma_{\eta_1}2\omega)} + \frac{B_2'}{(\omega_P^2 - 4\omega^2 + i\Gamma_P 2\omega)(\omega_{\eta_2}^2 - 4\omega^2 + i\Gamma_{\eta_2}2\omega)} \tag{63}$$

Figure 2b of this Supplementary Information shows that this latter equation fits rather well the MD data for $\beta'(\omega,\omega)$ (apart from the two peaks existing at frequencies of ω =90 GHz and 267 GHz, which may be numerical residuals of $\beta(0,\omega)$). Such overall agreement further validates our analytical model, that also results in Eq. (43).

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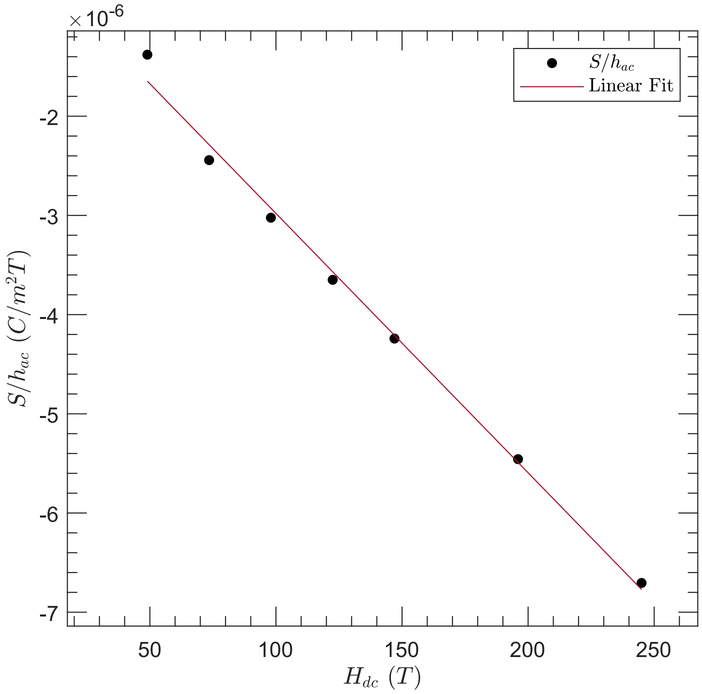

FIG. 1. (Color online)

Behavior of $\frac{S}{h_{ac}}$ as a function of the dc magnetic field when $h_{ac} = 61.2\,T$ and $\omega = 160\,GHz$, as obtained from MD simulations

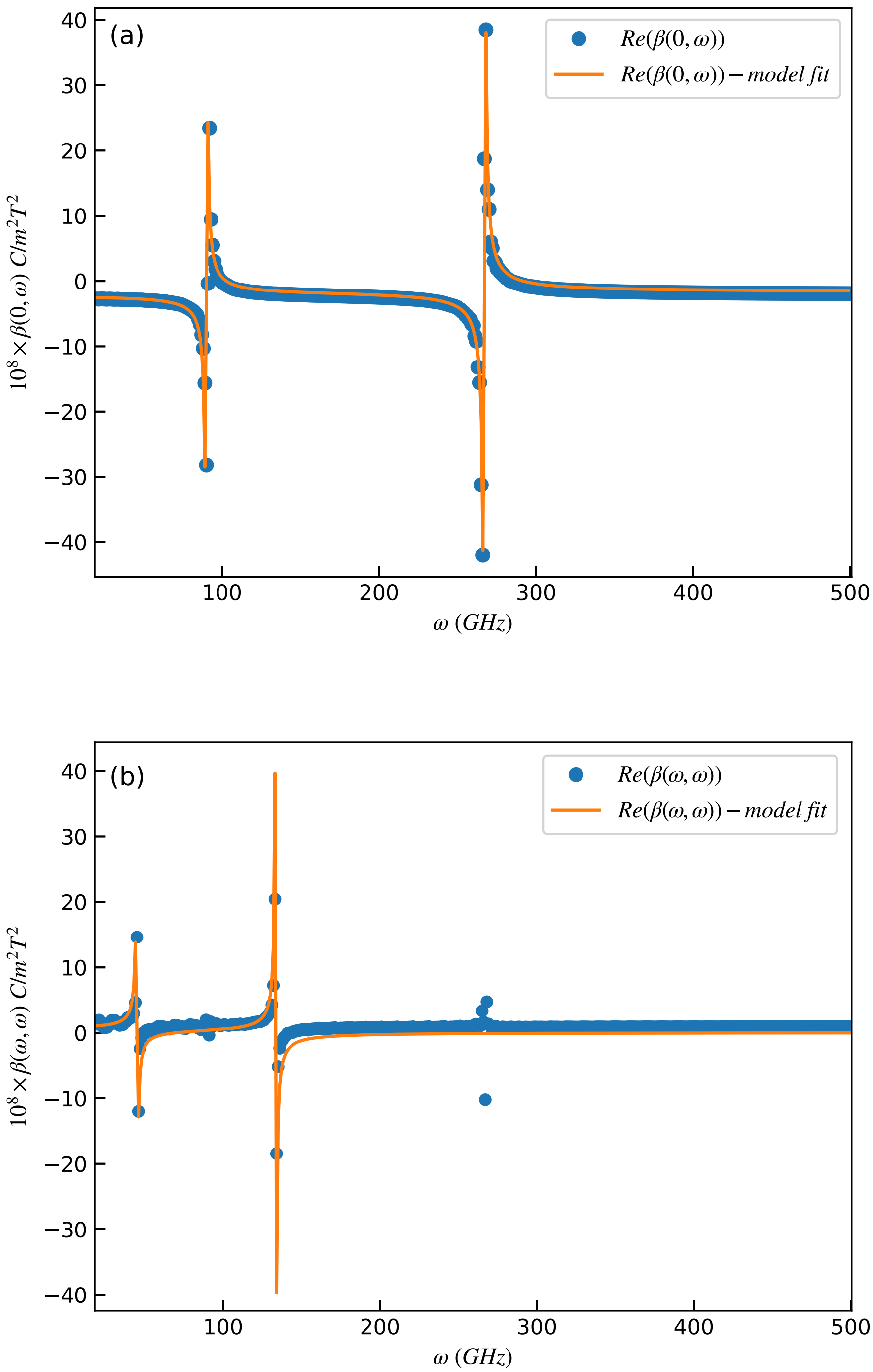

FIG. 2. (Color online)

The results obtained from MD simulations and the fit generated based on the analytical model for the real part of (a) $\beta(0,\omega)$ and (b) $\beta(\omega,\omega)$