Supplementary Material to the article "Magnetic, dielectric and magnetoelectric phenomena at low-temperature magnetic transitions in $GdFeO_3$"

To analyze the observed phenomena in $GdFeO_3$ at phase transitions, we used the approach developed in [1, 2] to study low-temperature phenomena in orthoferrites, orthochromites with rare earth ions, and present the nonequilibrium thermodynamic potential as

$$\Phi(\mathbf{F}, \mathbf{G}, \boldsymbol{\sigma}_\alpha) = \Phi_{Fe}(\mathbf{F}, \mathbf{G}) - N\{f_x[\mu(H_x+aF_x)+ \lambda_{xz}G_z]+ f_y\mu(H_y+aF_y) + f_z[\mu(H_z+aF_z)+ \lambda_{zx}G_x]+ c_y\lambda_{yz}G_z+ c_z\lambda_{zy}G_y \}+ \langle H_{Gd\text{-}Gd}\rangle+\tfrac{1}{4}NT\sum_{\alpha=1}^{4}S(\sigma_\alpha) \quad \text{(S1)}$$

where **F**, **G** are the vectors of ferro- and antiferromagnetism of the Fe subsystem, $\boldsymbol{\sigma}_\alpha$ – are the magnetic moments of the Gd^{3+} ion (normalized to its magnetic moment μ) in one of the four nonequivalent positions (sublattices) α =1-4 in the unit cell, which are the order parameters of the rare- earth subsystem, as well as their symmetrized combinations $\mathbf{f}=(\boldsymbol{\sigma}_1+ \boldsymbol{\sigma}_2+ \boldsymbol{\sigma}_3+ \boldsymbol{\sigma}_4)/4$, $\mathbf{g}=(\boldsymbol{\sigma}_1- \boldsymbol{\sigma}_2+ \boldsymbol{\sigma}_3- \boldsymbol{\sigma}_4)/4$, $\mathbf{a}= (\boldsymbol{\sigma}_1- \boldsymbol{\sigma}_2- \boldsymbol{\sigma}_3+ \boldsymbol{\sigma}_4)/4$, $\mathbf{c}= (\boldsymbol{\sigma}_1+ \boldsymbol{\sigma}_2- \boldsymbol{\sigma}_3- \boldsymbol{\sigma}_4)/4$. The first term in (1) represents the thermodynamic potential of the Fe subsystem

$$\Phi_{Fe}(\mathbf{F}, \mathbf{G}) = \tfrac{1}{2}\Lambda\mathbf{F}^2-d(F_xG_z-F_zG_x) - M_0\mathbf{FH}+\tfrac{1}{2}K_{ac}G_z^{\,2}+\tfrac{1}{2}K_{ab}G_y^{\,2} +\ldots \quad \text{(S2)}$$

where Λ, d are constants of isotropic and antisymmetric Fe-Fe exchange, respectively, $K_{ac} >0$, $K_{ab}>0$ are magnetic anisotropy constants that stabilize the high-temperature phase $\Gamma_4(F_zG_x)$, M_0 – is the magnetic moment of Fe^{3+} ions. The second term describes the interaction of Gd^{3+} ions with an external magnetic field **H** and Gd-Fe interaction, which includes isotropic (a) and anisotropic (λ_{ij}) Gd-Fe exchange [3], $\mu = \mu_B g_J S_{Gd} =7\mu_B$ is the magnetic moment of Gd^{3+}, N is the number of Gd^{3+} ions. The last two terms in (1) are determined, respectively, by the contributions of the Gd-Gd interaction

$$\langle H_{Gd\text{-}Gd}\rangle=-N[\tfrac{1}{2}\lambda^{ff}_{xx}f_x^{\,2}+\tfrac{1}{2}\lambda^{ff}_{yy}f_y^{\,2}+\tfrac{1}{2}\lambda^{ff}_{zz}f_z^{\,2}+\tfrac{1}{2}\lambda^{cc}_{xx}c_x^{\,2}+\tfrac{1}{2}\lambda^{cc}_{yy}c_y^{\,2}+\tfrac{1}{2}\lambda^{cc}_{zz}c_z^{\,2}+\lambda^{fc}_{xx}f_xc_y+\lambda^{cf}_{yy}f_yc_x +\tfrac{1}{2}\lambda^{gg}_{xx}g_x^{\,2}+\tfrac{1}{2}\lambda^{gg}_{yy}g_y^{\,2}+\tfrac{1}{2}\lambda^{gg}_{zz}g_z^{\,2}+\tfrac{1}{2}\lambda^{aa}_{xx}a_x^{\,2}+\tfrac{1}{2}\lambda^{aa}_{yy}a_y^{\,2}+\tfrac{1}{2}\lambda^{aa}_{zz}a_z^{\,2}+\lambda^{ga}_{xy}g_xa_y+\lambda^{ga}_{yx}g_ya_x], \quad \text{(S3)}$$

and the entropy of the Gd subsystem

$$S(\sigma_\alpha)=\ln(2S+1)-\int_0^{\sigma_\alpha}B_S^{-1}(\sigma)d\sigma, \quad \text{(S4)}$$

The values λ^{ff}_{xx}, λ^{ff}_{yy} ... are the constants of the Gd-Gd interaction, and $B_S^{-1}(\sigma)$ is the inverse Brillouin function of the Gd spins which is described in the molecular field approximation.

Minimizing the thermodynamic potential (S1) with respect to **F**, taking into account **FG**=0 due to saturation of the Fe sublattices and F<<G≈1, we find

$$\mathbf{F} = [\mathbf{H}_t - \mathbf{G}(\mathbf{H}_t\mathbf{G})]/\Lambda, \quad \text{(S5)}$$

and exclude it from the thermodynamic potential, where $\mathbf{H}_t = (M_0H_x + dG_z +N\mu af_x, M_0H_y + N\mu a f_y, M_0H_z - dG_x +N\mu af_z)$. Further, by minimizing (S1) with respect to the Gd order parameters $\boldsymbol{\sigma}_\alpha$ (or their basis vectors **f**, **g**, **a**, **c**) we get for them a system of molecular field equations

$$\boldsymbol{\sigma}_\alpha = (\mathbf{h}_\alpha/h_\alpha)B_s(h_\alpha/k_BT), \quad \text{(S6)}$$

where the effective fields at the four positions α of Gd ions are equal

$$\mathbf{h}_\alpha = n_\alpha^{\,f}\,\mathbf{h}_f + n_\alpha^{\,c}\,\mathbf{h}_c + n_\alpha^{\,g}\,\mathbf{h}_g + n_\alpha^{\,a}\,\mathbf{h}_a\ , \quad \text{(S7)}$$

in which

$$\mathbf{h}_f=\begin{pmatrix} \tilde{\mu}H_x+\tilde{\lambda}_{xz}G_z-\delta(\mu\vec{H}\vec{G})G_x+\lambda^{ff}_{xx}f_x+\lambda^{fc}_{xy}c_y+\Delta\lambda[f_x-(\vec{f}\vec{G})G_x] \\ \tilde{\mu}H_y-\delta(\mu\vec{H}\vec{G})G_y+\lambda^{ff}_{yy}f_y+\lambda^{fc}_{yx}c_x+\Delta\lambda[f_y-(\vec{f}\vec{G})G_y] \\ \tilde{\mu}H_z+\tilde{\lambda}_{zx}G_x-\delta(\mu\vec{H}\vec{G})G_z+\lambda^{ff}_{xx}f_z+\lambda^{fc}_{xy}c_y+\Delta\lambda[f_z-(\vec{f}\vec{G})G_z] \end{pmatrix}, \quad \text{(S8)}$$

$$\mathbf{h}_c=\begin{pmatrix}\lambda^{cc}_{xx}c_x+\lambda^{fc}_{yx}f_y\\ \lambda_{yz}G_z+\lambda^{cc}_{yy}c_y+\lambda^{fc}_{xy}f_x\\ \lambda_{zy}G_y+\lambda^{ff}_{zz}c_z\end{pmatrix},\ \mathbf{h}_g=\begin{pmatrix}\lambda^{gg}_{xx}g_x+\lambda^{ga}_{xy}a_y\\ \lambda^{gg}_{yy}g_y+\lambda^{ga}_{yx}a_x\\ \lambda^{gg}_{zz}g_z\end{pmatrix},\ \mathbf{h}_a=\begin{pmatrix}\lambda^{aa}_{xx}a_x+\lambda^{ga}_{yx}g_y\\ \lambda^{aa}_{yy}a_y+\lambda^{ga}_{xy}g_x\\ \lambda^{gg}_{zz}a_z\end{pmatrix}$$

represent the symmetrized components defining the corresponding basis vectors of Gd subsystem, and n_α^f=(1,1,1,1), n_α^c=(1,1,-1,-1), n_α^g=(1,-1,1,-1), n_α^c=(1,1,-1,-1), n_α^a=(1,-1,-1,1) specify combinations of these components in specific positions. In the above expressions $\widetilde{\lambda}_{xz}=\lambda_{xz}+\mu ad/\Lambda$, $\widetilde{\lambda}_{zx}=\lambda_{zx}-\mu ad/\Lambda$, $\widetilde{\mu}=\mu(1+\delta)$, where $\delta=M_0 a/\Lambda$ determines a small renormalization of the Gd^{3+} magnetic moment (also depending on **G**) due to the isotropic Gd-Fe exchange, $\Delta\lambda=N\mu^2a^2/\Lambda$ gives a similar correction to the parameters of the Gd-Gd interaction.

Taking into account that in the magnetic configuration $\Gamma_5(g_xa_y)$, Gd spins lie practically along the *a*-axis according to [4, 5] and our data in Fig.1a, meaning that the main Gd order parameter is the basic vector **g**, i.e. its g_x component, while a_y does not manifest itself in any way, we will further neglect the vector **a**, which is determined by the non-diagonal components of the Gd-Gd interaction λ^{ga}_{xy}.

For $H||c$ the system has a $\Gamma_4(G_xF_z)$ magnetic configuration for Fe and $\Gamma_{45}(f_zg_xa_y)$ for Gd subsystems, respectively. At Gd antiferromagnetic ordering the f_z и g_x order parameters according to Eq.(S6), are determined by the equations:

$$f_z=(\widetilde{\mu}H_z+\widetilde{\lambda}_{zx}G_x)/(\lambda^{gg}_{xx}-\widetilde{\lambda}^{ff}_{zz}),\quad g_x^2=\sigma_0^2-f_z^2,\qquad a_y\approx 0 \qquad \text{(S9)}$$

Where σ_0 satisfies the equation $\sigma_0=B_S(\sigma_0\lambda^{gg}_{xx}/k_BT)$. In this phase the Gd f_z ferromagnetic component linearly depends on the external and exchange $\widetilde{\lambda}_{zx}G_x$ fields with a constant, i.e. temperature-independent, slope, like transverse susceptibility in antiferromagnets. The region of existence of this phase is limited by the magnitude of the Gd spin-flip transition field $H_z^{s\text{-}flip}(T)$, at which g_x vanishes in (S9) $\widetilde{\mu}H_z^{s-flip}(T)=(\lambda^{gg}_{xx}-\widetilde{\lambda}^{ff}_{zz})\sigma_0(T)-\widetilde{\lambda}_{zx}G_x$ and which becomes zero at the Neel point (a small shift due to a small exchange field is omitted here). At low temperatures for T→0, the field $H_z^{s-flip}(0)$ can be estimated from the magnetization curve along the c-axis (Fig. 2a) as the intersection point of the linear part of the magnetization curve and its saturation value, which gives a field ~3.4 T, being noticeably higher than that we observed at 1.85 K.

In fields above the spin-flip transition, the remaining f_z component is determined by the equation $f_z=B_S((\widetilde{\mu}H_z+\widetilde{\lambda}_{zx}G_x+\widetilde{\lambda}^{ff}_{zz}f_z)/k_BT)$, in which H_z makes the main contribution to the effective field and the magnetic moment of gadolinium is saturated. The direction of the Gd-Fe exchange field below the compensation point coincides with the external field, i.e. $\widetilde{\lambda}_{zx}G_x>0$. In this case, the spontaneous weakly ferromagnetic Fe moment due to the Dzyaloshinsky-Moriya interaction, is $m_0^{Fe}G_x\equiv-M_0(d/\Lambda)G_x<0$, i.e. it lies against the external field and its Zeeman contribution $m_0^{Fe}G_xH_z$ to the thermodynamic potential increases linearly with increasing H_z. Since an energy of the Gd moment in the exchange Gd-Fe field is saturated in large fields, this leads to the change sign of G_x dependent part of the thermodynamic potential

$$\Delta\Phi(G_x)=G_x(m_0^{Fe}H_z-N\widetilde{\lambda}_{zx}f_z) \qquad \text{(S10)}$$

at some field $H_z\equiv H_{rev}=N\widetilde{\lambda}_{zx}f_z(H_z,T)/m_0^{Fe})$, where $f_z\to 1$, when $T\to 0$. This causes a reorientation (sign change) of $G_x\to -G_x$, accompanied by a change in the spontaneous canting of the Fe sublattices from the direction against field to the direction along one and an increase in the magnetic moment at low temperatures by $\sim 2|m_0^{Fe}|$. In this case, the Gd-Fe exchange coupling, determined by the term $-N\widetilde{\lambda}_{zx}G_xf_z$ in (S10), increases, i.e. the energetically preferable mutual orientation (coupling) of the Gd and Fe moments are violated. The temperature dependence of the transition field between these states can be represented as (for a small exchange Gd-Fe interaction $\widetilde{\lambda}_{zx}$, true in our case)

$$k_B T = \frac{(\tilde{\mu} H_z^0 + \tilde{\lambda}_{zz}^{ff}) H_z / H_z^0}{B_S^{-1}(H_z / H_z^0)} \tag{S11}$$

where $H_z^o = N\tilde{\lambda}_{zx} / m_0^{Fe}$ is the transition field at T=0. As the temperature increases, the magnitude of the transition field decreases and vanishes at the compensation point $k_B T_{comp} = (N\tilde{\mu}\tilde{\lambda}_{zx} / m_0^{Fe} + \tilde{\lambda}_{zz}^{ff})(S_{Gd}+1)/3S_{Gd} \equiv (\tilde{\mu} H_z^0 + \tilde{\lambda}_{zz}^{ff})(S_{Gd}+1)/3S_{Gd}$. It can be seen that T_{comp} is related to $H_z^0$and the paramagnetic Curie temperature of the Gd subsystem $k_B \Theta_c = -\tilde{\lambda}_{zz}^{ff}(S_{Gd}+1)/3S_{Gd}$. According to our data (Fig.2a), the value of H_z^0=3.5-3.8 T and gives a value of T_{comp} close to the observed one. Thus, at the $H||c$ axis, the phase H-T diagram includes the spin-flip transition line in the Gd subsystem $H_z^{s\text{-}flip}(T)$ and the $H_{rev}(T)$ line corresponding to reversal of spontaneous Fe weak ferromagnetic moment.

Let us now turn to electrical polarization and dielectric constant. Polarization $P_z = -\partial\Phi/\partial E_z = [\lambda_1 G_x + \lambda_1' f_z + \lambda_1'' H_z] g_x$ exists only below antiferromagnetic Gd ordering and is determined mainly by the temperature and field dependence of the main order parameter $g_x(T, H_z)$.

The electrical susceptibility χ_z^E (or dielectric constant $\varepsilon_z = 1+4\pi\chi_z^E$) is determined, in addition to the usual lattice contribution $(\chi_z^E)_{lat}$, also by magnetoelectric contributions from the Gd order parameters dg_x/dE_z and df_z/dE_z, depending on E_z

,
$$\chi_z^E = dP_z / dE_z = -\Phi_{E_z E_z} - \Phi_{E_z g_x} dg_x / dE_z - \Phi_{E_z f_z} df_z / dE_z \tag{S12}$$

where $\Phi_{E_z E_z} = \partial^2\Phi / \partial E_z^2$, $\Phi_{E_z g_x} = \partial^2\Phi / \partial E_z \partial g_x$ etc. are partial derivatives of the total nonequilibrium potential Φ, where the magnetoelectric part (1) and the lattice part are added. The values dg_x/dE_z and df_z/dE_z are determined by differentiating by E_z the equations $\partial\Phi/\partial g_x$=0 and $\partial\Phi/\partial f_z$=0 for the equilibrium values of the order parameters g_x and f_z corresponding to the minimum Φ. As a result, one gets for χ_z^E

$$\chi_z^E = (\chi_z^E)_{\text{lat}} + (\Phi_{E_z g_x}^2 \Phi_{g_x g_x} + \Phi_{E_z f_z}^2 \Phi_{f_z f_z} - 2\Phi_{E_z g_x} \Phi_{E_z f_z} \Phi_{g_x f_z}) / \Delta \tag{S13}$$

where value $\Delta = \Phi_{g_x g_x} \Phi_{f_z f_z} - \Phi_{g_x f_z}^2 \geq 0$ determines the stability of the Gd magnetic configurations, respectively, $\Gamma_{45}(f_z\ g_x a_y)$ at $T<T_N(H_z)$ and $\Gamma_4(f_z)$ at $T>T_N(H_z)$. At the phase transition, when Δ→0, the electrical susceptibility increases. In the $\Gamma_4(f_z)$ phase, where g_x =0, the susceptibility is

$$\chi_z^E = (\chi_z^E)_{\text{lat}} + \frac{(\lambda_1 G_x + \lambda_1' f_z + \lambda_1'' H_z)^2}{N[(\tilde{\mu} H_z + \tilde{\lambda}_{zx} G_x + \tilde{\lambda}_{zz}^{ff} f_z)/f_z - \lambda_{xx}^{gg}]}, \tag{S14}$$

where the displacement of the Neel point in the field is taken into account, and $f_z = B_S((\tilde{\mu} H_z + \tilde{\lambda}_{zx} G_x + \tilde{\lambda}_{zz}^{ff} f_z)/k_B T)$ is determined by the molecular field equation obtained from (S6). For small fields, it is reduced to the expression

$$\chi_z^E = (\chi_z^E)_{\text{lat}} + (\lambda_1 G_x + \lambda_1' f_z + \lambda_1'' H_z)^2 (S_{Gd}+1)/3S_{Gd}(T - T_N) k_B N, \tag{S15}$$

clearly demonstrating the divergence of electrical susceptibility at the Neel point. In Fig. 4a, b are the calculated temperature and field dependences of the dielectric constant and polarization, which are generally consistent with the experiment.

In the $H||b$ field, which, like $H||c$, suppresses the antiferromagnetic ordering of Gd and does not change the orientation of the vector **G**, electric polarization and permittivity behave similarly to the case of $H||c$. This indicates that the magnetoelectric contributions $\lambda_1' g_x f_z$ and $\lambda_1'' g_x H_z$ in Eq.(1) associated with the magnetization of Gd along the c axis are apparently small.

Let us now consider the case of the $H||a$ axis, when the magnetic field induces a spin-flop transition in the Gd subsystem $\Gamma_{45}(g_x f_z) \rightarrow \Gamma_{27}(g_z f_x c_y)$ at low temperatures accompanied by a spin reorientation in the Fe subsystem. To analyze the behavior of the magnetic structure, we specify the thermodynamic potential. In fields small compared to the spin-flip transition field in the Gd subsystem, when the ferromagnetic, ***f*** and antiferromagnetic, ***c*** order parameters are small compared to **g**, the nonequilibrium thermodynamic potential can be minimized with respect to ***f*** and ***c*** and represented as a function only of **g** and **G** orientation in the *xz* plane (*ac*):

$$\Phi(\vec{G},\vec{g}) \approx -\tfrac{1}{2}\chi_{\perp}^{Gd}\vec{H}_f^2 + \tfrac{1}{2}\Delta\chi^{Gd}(\vec{H}_f\vec{g})^2/g^2 - \tfrac{1}{2}\chi_{\perp}^{Gd}\vec{H}_c^2 + \tfrac{1}{2}K_{ac}^{Gd}g_z^2 + \tfrac{1}{2}(K_{ac}-\chi_{\perp}^{Fe}H_x^2)G_z^2 - m_0^{Fe}H_xG_z \tag{S16}$$

where $\vec{H}_f = (H_x + H_x^{eff}G_z, 0, H_z^{eff}G_x)$, $\vec{H}_c = (0, H_y^{eff}G_z, 0)$ are the effective fields on Gd ions magnetizing them by ***f*** or ***c*** type, $H_{x,z}^{eff} = \tilde{\lambda}_{xz,zx}/\tilde{\mu}$, $H_y^{eff} = \lambda_{yz}/\tilde{\mu}$; $K_{ac}^{Gd} = N(\lambda_{xx}^{gg} - \lambda_{zz}^{gg}) > 0$ is the anisotropy energy of the Gd subsystem; $\chi_{\perp}^{Gd} = N\mu^2/(\lambda^{gg}-\lambda^{ff})$ and $\chi_{\parallel}^{Gd} = \chi_{\perp}^{Gd}(\lambda^{gg}-\lambda^{ff})B_S'/[(\lambda^{gg}-\lambda^{ff})B_S' + g^2k_BT]$ are Gd transverse and longitudinal susceptibility, respectively, while $\Delta\chi^{Gd} = \chi_{\perp}^{Gd} - \chi_{\parallel}^{Gd}$ is their difference, and $\chi_{\perp}^{Fe} = M_0^2/\Lambda$ is the transverse susceptibility of the Fe subsystem. The Eq.(S16), where the terms dependent on g^2 were omitted, describes two exchange coupled antiferromagnetic subsystems in the magnetic field H_x. As the analysis shows, the stable state is $G_x \approx g_x \approx \pm 1$ for small fields , while with H_x increasing (but remaining $H_x \ll H_{s\text{-}flip}$) a spin–flop phase $g_z \approx 1$ for Gd ions and an angular structure $0<G_z<1$ (Γ_{24}) in the Fe subsystem become stable. With further H_x growth the **G** vector turns to the c-axis until it completely switches to the $\Gamma_2(G_zF_x)$ state. The first order phase transition occurs between the phases at the field satisfying of equality of their free energy

$$H_{x1} \approx \sqrt{(K_{ac}^{Gd} + \chi_{\perp}^{Gd}H_z^{eff2})/(\Delta\chi^{Gd} + \chi_{rot}^{Fe})} \tag{S17}$$

where $\chi_{rot}^{Fe} \approx (m_0^{Fe} + m_x^{Gd})^2/\tilde{K}_{ac}^{Fe}$ is the susceptibility of Fe spin rotation, $\tilde{K}_{ac}^{Fe} = K_{ac}^{Fe} - \chi_{\perp}^{Gd}(H_x^{eff2} + H_y^{eff2} - \chi_{\perp}^{Fe}H_x^2)$ is Fe effective anisotropy energy in the *ac* plane, and $m_x^{Gd} = \chi_{\perp}^{Gd}H_x^{eff}$ is the Gd contribution to the weak ferromagnetic moment. The orientation of the Fe spins in the angular Γ_{24} phase is characterized approximately the projection $G_z = \cos\theta \approx (m_0^{Fe} + m_x^{Gd})H_x/(\tilde{K}_{ac}^{Fe} - \chi_{\perp}^{Fe}H_x^2)$, which increases with the H_x growth and approaches unity at the end of the reorientation. Another scenario of field-induced spin reorientation suggests a spin-flop transition simultaneously in Gd ($g_zf_x \rightarrow f_x$) and Fe ($G_x \rightarrow G_z$) subsystems at threshold field

$$H_{x2} \approx \frac{m_0^{Fe} + m_x^{Gd}}{\chi_{\perp}^{Fe} + \chi_{\perp}^{Gd} - \chi_{\parallel}^{Gd}}\left[\sqrt{1 + \frac{K_{ac}^{eff}(\chi_{\perp}^{Fe} + \chi_{\perp}^{Gd} - \chi_{\parallel}^{Gd})}{(m_0^{Fe} + m_x^{Gd})^2}} - 1\right], \tag{S18}$$

where $K_{ac}^{eff} = K_{ac}^{Fe} + K_{ac}^{Gd} - \chi_{\perp}^{Gd}(H_x^{eff2} + H_y^{eff2} - H_z^{eff2})$ is the effective anisotropy energy in the *ac* plane. Using the known data for the parameters of the Gd-Fe interaction [3], as well as our magnetization data to determine $\chi_{\perp}^{Gd}$, K_{ac}^{Gd} , we simulated the temperature dependences of the threshold fields of spin-reorientation and spin-flip transitions (Fig.6) for $\chi_{\perp}^{Gd}$=4.6 10^{-3} cm^3/g, $\chi_{\perp}^{Fe}$=0.9 10^{-4} cm^3/g, K_{ac}^{Gd}=0.8 10^5erg/g, K_{ac}^{Fe}=1.3 10^5erg/g, m_0^{Fe}=1.2 emu/g, H_x^{eff}=1 kOe, H_y^{eff}=4 kOe, H_z^{eff}=-0.5 kOe, *a*=-180 kOe, λ_f=-12.5 K. The calculated threshold fields are consistent with the experiment above and below the Neel point (Fig.6), including data from [3], and indicate that the spin reorientation can occur either through an angular phase in the Fe subsystem or directly to the $\Gamma_2(G_zF_x)$ phase. The both scenario are discussed in the main text of the paper.

In the fields above the spin-flop, there is a small break in the magnetization curves (and its derivative) during the spin-flip transition to Gd ($g_zf_x \rightarrow f_x$), and a peak is observed in the field dependence of the dielectric constant $\varepsilon_z(H_x)$ when g_z is suppressed of approximately the same magnitude as in the field along a- and b-axes which suppress g_x-component. This indicates that the magnetoelectric constants λ_1 and λ_2 in (1) are approximately the same magnitude. This is also confirmed by the small dielectric anomaly during the spin-flop transition on the lower branch 1 of the curve $\varepsilon_z(H_x)$ (Fig.5a).

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