Reply to "Comment on 'Ultrafast polarization switching in ferroelectrics' "

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Recently, a method of ultrafast polarization switching in ferroelectrics has been suggested. The basic idea of the method is to employ the effect of self-acceleration of polarization dynamics due to a resonator feedback field. This is the idea of principle whose efficiency is demonstrated by an Ising-type model in a transverse field. Of course, the practical realization of the method requires the choice of appropriate materials, which is a separate problem. For example, the standard order-disorder ferroelectrics with spatially symmetric double-well potentials cannot be used for this purpose since, because of the symmetry, they lack the transverse polarization. However, ferroelectrics with asymmetric potentials, possessing this polarization, can be used. Moreover, if the potential asymmetry, hence, the transverse polarization, could be regulated, e.g., by shear stress or shear strain, this could provide a tool for governing the process of polarization switching.

The main goal of the paper [1] has been to attract attention to the possibility of accelerating the polarization switching in ferroelectrics by using the self-acceleration effect caused by the action of a resonator-cavity feedback field. This is the idea of principle that, here, is considered for ferroelectrics. It goes without saying that the method is not necessarily applicable to any particular material. Thus, in Ref. [2], it is mentioned that the standard order-disorder ferroelectrics with spatially symmetric double wells cannot be used for this purpose. Below, we explain that the model considered in Ref. [1] assumes the general case of asymmetric potentials for which the method is applicable. We also emphasize that, since there are various types of ferroelectric systems [3–6], there can exist other materials for which the suggested idea could work.

Of course, to illustrate the idea, one has to consider some model. We considered an Ising-type model in a transverse field. This kind of model, employing the spin representation, is widely used for order-disorder ferroelectrics [3–6] and relaxor ferroelectrics [7].

In order that the suggested method could be realized, the existence of two spin components of polarization, longitudinal and transverse, is required. If one keeps in mind an order-disorder ferroelectric with lattice-site double wells that are ideally symmetric with respect to spatial inversion (especially with respect to the inversion $x \rightarrow -x$), then, there is only a longitudinal component, and the sample polarization is expressed through the z component of the spin operator. But, in the general case of an asymmetric potential, the sample polarization contains a term with the x component of spin. The asymmetry can be induced by shear stress or strain or by incorporating into the sample admixtures or vacancies. Thus, the inclusion of vacancies in order-disorder ferroelectrics is attributed to the breaking of spatial inversion symmetry along different directions [8]. The symmetry in order-disorder ferroelectrics can be distorted by the action of a transverse electric field [9].

In the paper [1], the general case of an asymmetric site potential is treated when the expression for polarization possesses both spin components, longitudinal and transverse. In order to be explicit, let us briefly show how these components arise.

The most general approach requires to start from the microscopic Hamiltonian in the second-quantization representation,

$$\hat{H} = \int \psi^{\dagger}(\mathbf{r}) H_1(\mathbf{r}) \psi(\mathbf{r}) \, d\mathbf{r} - \hat{\mathbf{P}} \cdot \mathbf{E}_{\text{tot}}$$
$$+ \frac{1}{2} \int \psi^{\dagger}(\mathbf{r}) \psi^{\dagger}(\mathbf{r}') \Phi(\mathbf{r} - \mathbf{r}') \psi(\mathbf{r}') \psi(\mathbf{r}) d\mathbf{r} \, d\mathbf{r}'. \quad (1)$$

Here, ψ's are field operators, the site Hamiltonian is

$$H_1(\mathbf{r}) = -\frac{\nabla^2}{2m} + U(\mathbf{r}), \quad (2)$$

the potential $U(\mathbf{r})$, generally, is not symmetric with respect to the spatial inversion $\mathbf{r} \rightarrow -\mathbf{r}$, Φ is the interaction potential, and E_{tot} is an external electric field acting on the polarization operator,

$$\hat{\mathbf{P}} = \int \psi^{\dagger}(\mathbf{r}) \mathbf{P}(\mathbf{r}) \psi(\mathbf{r}) d\mathbf{r}, \quad (3)$$

where the local polarization $\mathbf{P}(\mathbf{r})$ is caused by the charge distribution satisfying the condition of the sample neutrality.

Considering an insulating sample where particles are localized in the vicinity of the lattice sites, the field operators

can be expanded over localized orbitals, for example, over the well-localized Wannier functions [10],

$$\psi(\mathbf{r}) = \sum_{nj} c_{nj} w_n(\mathbf{r} - \mathbf{r}_j). \qquad (4)$$

For the unity filling factor, the no-double-occupancy condition is valid: $\sum_n c_{nj}^\dagger c_{nj} = 1$ and $c_{mj} c_{nj} = 0$. For an insulating lattice, the no-hopping condition is satisfied: $c_{mi}^\dagger c_{nj} = \delta_{ij} c_{mj}^\dagger c_{nj}$.

One assumes that the most populated are the lowest two energy levels, whereas the other levels can be neglected. This allows one to introduce the spin representation,

$$c_{1j}^\dagger c_{1j} = \tfrac{1}{2} + S_j^x, \quad c_{2j}^\dagger c_{2j} = \tfrac{1}{2} - S_j^x,$$
$$c_{1j}^\dagger c_{2j} = S_j^z - i S_j^y, \quad c_{2j}^\dagger c_{1j} = S_j^z + i S_j^y. \qquad (5)$$

Thus, omitting nonoperator terms, we come to the Hamiltonian,

$$\hat{H} = -\sum_j \left(\Omega_j S_j^x - H_j S_j^z \right) - \sum_j \hat{\mathbf{P}}_j \cdot \mathbf{E}_{\text{tot}}$$
$$+ \frac{1}{2} \sum_{i \neq j} \left(B_{ij} S_i^x S_j^x - J_{ij} S_i^z S_j^z \right), \qquad (6)$$

in which $\Omega_j \equiv H_{jj}^{22} - H_{jj}^{11} + \frac{1}{2} \sum_i C_{ij}$, $H_j \equiv H_{jj}^{12} + H_{jj}^{21}$, $B_{ij} \equiv V_{ij}^{1111} + V_{ij}^{2222} - 2V_{ij}^{1221}$, $C_{ij} \equiv V_{ij}^{2222} - V_{ij}^{1111}$, and $J_{ij} \equiv -4V_{ij}^{1122}$ with H_{ij}^{mn} being the matrix elements of Hamiltonian (2) over the Wannier functions and V_{ij}^{mnkl}, the corresponding matrix elements of the interaction potential. The polarization operator takes the form

$$\hat{\mathbf{P}}_j \equiv \mathbf{d}_0 S_j^z + \mathbf{d}_1 S_j^x, \qquad (7)$$

where $\mathbf{d}_0 \equiv \mathbf{P}_{12} + \mathbf{P}_{21}$, $\mathbf{d}_1 \equiv \mathbf{P}_{11} - \mathbf{P}_{22}$, and

$$\mathbf{P}_{mn} \equiv \int w_m^*(\mathbf{r} - \mathbf{r}_j) \mathbf{P}(\mathbf{r}) w_n(\mathbf{r} - \mathbf{r}_j) d\mathbf{r}.$$

The total external electric field contains a longitudinal and a transverse components $\mathbf{E}_{\text{tot}} = E_0 \mathbf{e}_z + E \mathbf{e}_x$. Here, E_0 is a fixed external field, and E is the feedback field of a resonant cavity.

The term H_j can be included into the field E_0. The magnitude of the interaction parameter B_{ij} is usually much smaller than the tunneling frequency Ω_j that can be taken the same

for all lattice sites $\Omega_j = \Omega$. Omitting the term with B_{ij} is not principal since it is easy to show [11] that its main role is the renormalization of the frequency Ω_j. As a result, we obtain the Hamiltonian,

$$\hat{H} = -\Omega \sum_j S_j^x - \frac{1}{2} \sum_{i \neq j} J_{ij} S_i^z S_j^z - \sum_j \hat{\mathbf{P}}_j \cdot \mathbf{E}_{\text{tot}}. \qquad (8)$$

When the potential configuration at each lattice site is symmetric with respect to spatial inversion (especially with respect to the inversion $x \to -x$) so that the density $|w_n(\mathbf{r})|^2$ is also symmetric with respect to the spatial inversion, then, the diagonal matrix elements $\mathbf{P}_{nn}$ are zero because of which the polarization operator contains only the longitudinal spin component $\mathbf{d}_0 S_j^z$. However, in the general case, when the potential relief is not inversion symmetric, the polarization operator contains both the longitudinal as well as the transverse spin components as in Eq. (7). This general case is assumed in Ref. [1].

Thus, in the general situation, the polarization operator possesses two spin components longitudinal and transverse. This is sufficient for realizing the effect of the self-acceleration of the polarization switching by the resonator feedback field, which is the main point of the paper [1]. It is useful to note that, in addition to different ferroelectrics [3–7] and multiferroics [12,13], ferroelectric-type spin models are widely used for describing the systems of polar molecules, Rydberg atoms, Rydberg-dressed atoms, dipolar ions, vacancy centers in solids, and quantum dots [14–19]. These systems can form self-assembled lattice structures or can be loaded into external potentials imitating crystalline models. The characteristics of these dipolar materials can be varied in a very wide range. Therefore, it looks feasible to find the appropriate material for the realization of the effect considered in the paper [1].

Moreover, the fact that the potential asymmetry leads to the appearance of the transverse polarization component containing S_j^x suggests the way of regulating the polarization switching. Thus, in a sample with symmetric wells where there is no transverse polarization, the longitudinal polarization can be frozen. By inducing the potential asymmetry, for instance, by subjecting the material to shear stress and shear strain, which would induce the transverse polarization, it would be possible to trigger the switching process.

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