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Correlation between dielectric and electro-optic properties related to domain dynamics in $RbHSeO_4$ crystals

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The variations of the effective electro-optic coefficient and of the dielectric permittivity as functions of the dc electric field are reported in rubidium hydrogen selenate at room temperature. Their frequency dispersions are measured in the range 20 Hz–1 MHz. The influence of the ac field amplitude on these properties is also studied. It is shown that the dielectric and the electro-optic responses are strongly nonlinear with the amplitude of the ac electric field, and that their frequency dispersions are closely linked to one another, up to the vicinity of the piezoelectric resonances. The role of domain dynamics in the dielectric and electro-optic properties is stressed. The domain contribution to the electro-optic response is explained by means of a simple model based on the tilt of the optical indicatrix in neighboring domains. [S0163-1829(98)02629-0]

I. INTRODUCTION

The fabrication and the study of materials with large electro-optical (EO) or nonlinear optical coefficients are always of a current interest. Among them, ferroelectric (FE) single crystals have been intensively investigated.[1] Indeed, in these materials EO properties are generally correlated to the linear dielectric susceptibility so that they are expected to increase in the vicinity of the paraelectric-ferroelectric phase transition, in connection with the maximum of the static permittivity, favoring large polarization fluctuations.[2] We have recently shown that the crystal of rubidium hydrogen selenate [$RbHSeO_4$ (RHSe)] possesses unusual large EO properties,[3] allowing low driving voltages for the modulation of light.

This crystal is known as a hydrogen-bonded ferroelectric with the Curie point at 371 K.[4] At room temperature RHSe presents the triclinic symmetry $P1$ and possesses ferroelastic, pyroelectric, and ferroelectric properties that have been widely studied.[5–7] Below T_C, the crystal always exhibits a layered domain structure with walls parallel to the (a,b) crystallographic planes, and the spontaneous polarization directed close to the b axis. Neighboring domains are each other symmetrical by a twofold rotation around the axis perpendicular to the (a,b) planes. This ferroelectric-ferroelastic (FEFEL) domain structure is especially soft—that means easily reversible and removable by a stress or by an electric field[8]—and it reappears spontaneously as soon as the field is released after saturation. This soft behavior is likely at the origin of some interesting properties of RHSe such as the large electro-optic effect,[3] deflection, and electrodeflection of light.[9] We have recently reported the influence of the dc field on the EO properties in recording the cycles of the birefringence versus increasing and decreasing electric field, and we have pointed out the role played by the domain structure in this large EO effect.[10] The aim of the present work is to correlate the EO properties with the dielectric response and thus to emphasize the role of domain dynamics in the properties of RHSe. Therefore, we have undertaken simultaneous measurements of the corresponding dielectric susceptibility and the EO coefficient as a function of the ac field, varying both in frequency and amplitude. We particularly emphasize the role of domain dynamics in the EO properties of RHSe in the frame of a simple model based on the tilting of the optical indicatrix caused by a field-induced reversal of the FEFEL domains.

II. EXPERIMENTAL

Dielectric and electro-optic measurements were carried out on the same RHSe sample that was cut from a solution-grown crystal and shaped as parallelepiped 2.6×2.0×6.1 mm^3. The smallest dimension d was cut approximately parallel to the ferroelectric b axis, and the largest one L—parallel to the propagation direction of the light in the experiments—was intentionally misoriented by 20° from the c axis in order to obtain a large EO effect.[10] The sample was electroded with silver paste on the b faces and placed into a closed cell filled with silicon oil to avoid damage due to moisture.

The EO experiments were done at the 633 nm wavelength of a He-Ne laser using the Sénarmont compensator setup described in Fig. 1, as well as the corresponding transmission factor I/I_0. The effective electro-optic coefficient r_{eff} under an ac field is deduced using the working point M_0, from the modulation factor $m=2J_\omega/I_0$, where J_ω is the amplitude of the optical signal modulated at the output of the analyzer and I_0 is the maximum transmitted intensity. The EO coefficient is derived from $n^3 r_{\mathrm{eff}}=(m\times\lambda\times d)/(\pi\times L\times V_m)$ where λ is the laser wavelength, and V_m is the amplitude of the applied ac voltage. The variation of r_{eff} is studied as a function of the frequency and amplitude of the ac field. This method differs from the procedure using the compensation of the phase shift

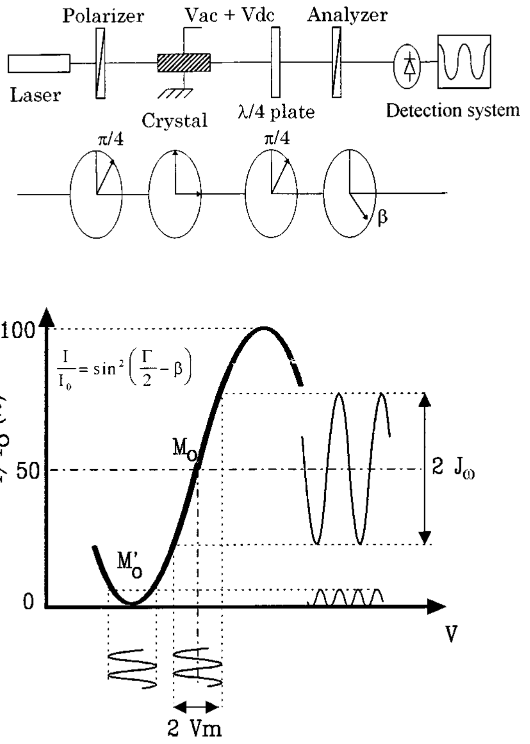

FIG. 1. Sénarmont's setup used for electro-optic and birefringence cycle measurements (upper part). Transmission factor corresponding to this optical arrangement showing two particular working points M_0 and M_0' (lower part).

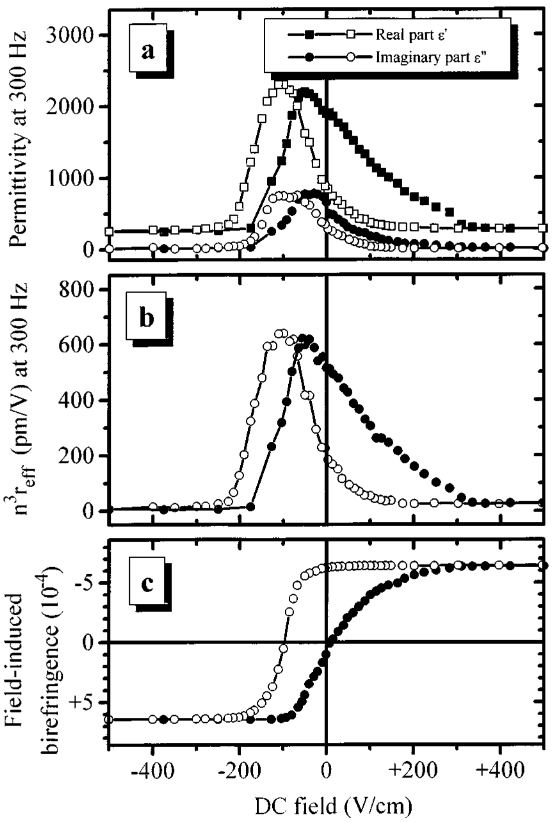

FIG. 2. dc cycles of the relative permittivity (a) and of the effective electro-optic coefficient (b) recorded under an ac electric field with a frequency of 300 Hz and an amplitude of 250 V/cm peak to peak. (c): cycle of the field-induced birefringence $\Delta n(E)-\Delta n_0$ recorded in the same configuration. Solid symbols: increasing bias. Open symbols: decreasing bias. The total cycling time of the experiments was approximately 4 h for (a) and (b), 2 h for (c). The solid lines are only guides to the eyes.

$\Gamma(E)=2\times\pi\times L\times\Delta n(E)/\lambda$, induced by a dc field. In this case the compensation is achieved step by step during dc cycling by appropriate rotations $\delta\beta=\delta\Gamma/2$ of the analyzer in order to track the extinction of the light—or the double frequency point M_0' (see Fig. 1) of the optical modulation if an ac field is superimposed as a probe. This method is employed to determine the birefringence cycles and the static value of the EO coefficient.

Dielectric measurements at high ac fields (from 10 to 350 V/cm peak-to-peak) were carried out using a Sawyer-Tower bridge, simultaneous to the EO measurements. Dielectric measurements at low ac fields (from 0.6 to 28 V/cm peak-to-peak) were independently performed with a Hewlett-Packard 4192 A setup. The experimental errors are typically 5% for the dielectric measurements and 10% for the EO measurements. However, this latter error can be larger (20%) for the values of n^3r_{eff} measured under low ac fields (<50 V/cm peak-to-peak).

III. RESULTS

A. dc cycles

The permittivity cycle $\varepsilon(E)$ and the electro-optic cycle $n^3r_{\mathrm{eff}}(E)$ were recorded versus increasing and decreasing dc field, from −500 V/cm to +500 V/cm, with a superimposed ac field (300 Hz, 250 V/cm peak-to-peak). The birefringence cycle $\Delta n(E)$ was also recorded in the same configuration by rotating the analyzer in the Sénarmont's setup in order to track the double-frequency point of the EO modulation during dc cycling.[10] The results are plotted in Fig. 2. The hysteresislike behavior of the dielectric permittivity versus increasing and decreasing dc field is evidenced in Fig. 2(a). In both saturated states ($|E_{\mathrm{dc}}|>300$ V/cm) the value $\varepsilon_{\mathrm{sat}}$ of the real part ε' is equal to approximately 250, while the imaginary part is nearly zero. In these saturated states the FE domain structure of the sample is almost completely removed so that the dielectric permittivity is mainly due to ionic contributions. The contribution of domain dynamics is responsible for the hysteresislike behavior that appears for dc fields ranging approximately from −200 to +200 V/cm. The peak value of ε' ($\approx$2300) is obtained at decreasing bias for a dc field of approximately −100 V/cm. This particular state corresponds to a maximal efficiency of domain reversal under the alternative field, and it will be referred to as the coercive state in the following paragraphs. This value of the coercive field is extremely low compared to others FE materials. For this reason, RHSe can be considered as a particularly ''soft'' FE crystal.

It should be noticed that the permittivity cycle is not symmetrical: the maximum of ε' with increasing bias appears for a dc field of approximately −50 V/cm, a value that is not the opposite to this obtained with decreasing bias. Moreover, the dc field required to saturate the sample for increasing bias is slightly larger than for decreasing bias. This asymmetry in the hysteresislike behavior should be attributed to a preexisting framework in the domain structure of the sample, prob-

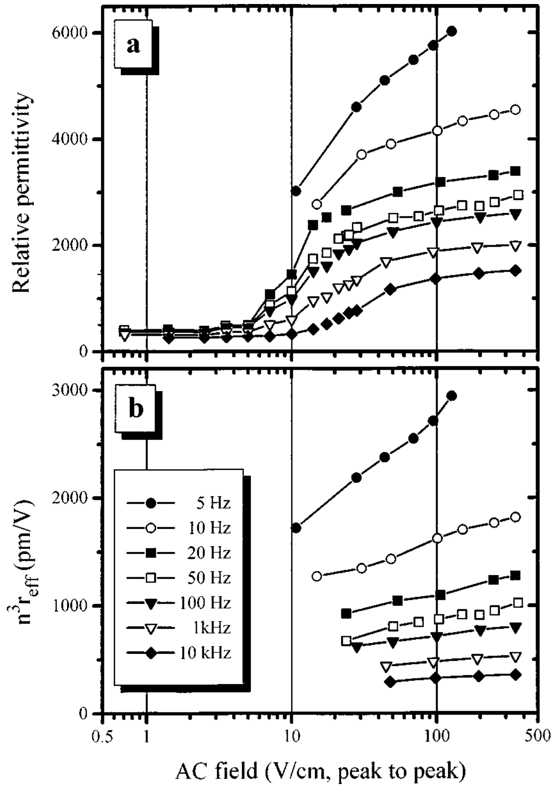

FIG. 3. Variations of the relative permittivity (a) and of the effective electro-optic coefficient (b) vs the amplitude of the ac field, in the coercive state of the sample. The solid lines are only guides to the eyes.

ably caused by defects acting as pinning points for domain walls inside the crystal or by preferential sites for domain nucleation at the electrodes. This asymmetry is not modified after several dc cycles.

As seen in Fig. 2(b), the effective electro-optic coefficient $n^3r_{\rm eff}$ of the sample exhibits the same hysteresislike behavior as the dielectric permittivity. This clearly shows that the large EO effect in RHSe is due to domain dynamics. Moreover, the EO coefficient $n^3r(E)$ is fairly proportional to the domain-related susceptibility, $\chi_d(E)=\varepsilon(E)-\varepsilon_{\rm sat}$. The EO cycle $n^3r(E)$ also looks proportional to the derivative of the birefringence cycle $\Delta n(E)$ that was recorded in the same configuration [Fig. 2(c)]. Unfortunately, it was not possible to record simultaneously the cycle of the ferroelectric polarization $P(E)$ because the cycling time during the experiments was too long (approximately 2 h).

B. Dependence of ε and $n^3r_{\rm eff}$ on the ac field amplitude

The dependences of both the permittivity and the electro-optic coefficient versus ac-field amplitude have been measured in the coercive state of the sample at several frequencies ranging from 5 Hz to 10 kHz. Results are reported in Fig. 3. Under low ac amplitudes ($E_{\rm ac}<5$ V/cm peak-to-peak) the dielectric response of polydomain RHSe [Fig. 3(a)] remains nearly linear and depends only slightly on the ac frequency. The value of this co-called ''initial permittivity'' fairly corresponds to the value $\varepsilon_{\rm sat}$ that is obtained in the fully saturated state [Fig. 2(a)], when the domain structure is nearly removed by the dc field. This indicates that the contribution of domain dynamics to the dielectric susceptibility of RHSe is weak under low ac field amplitudes below 5 V/cm.

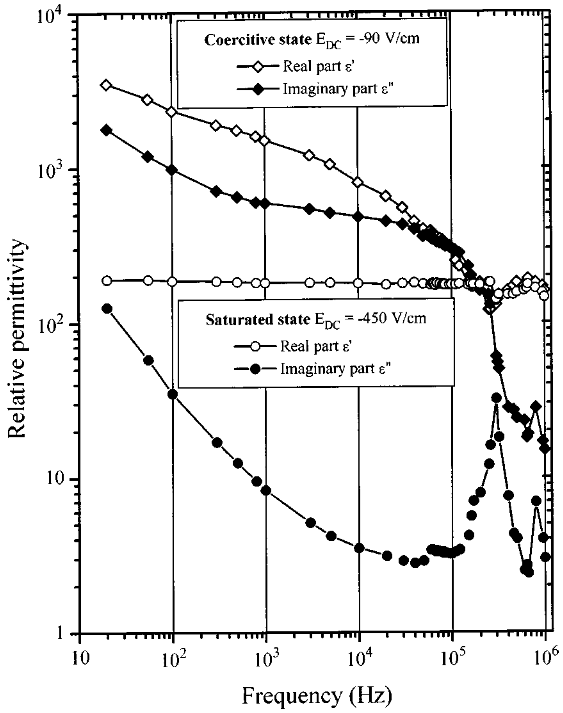

FIG. 4. Dispersion of the relative dielectric permittivity and losses vs frequency recorded in the crystal in the coercive state (squares) and in the saturated state (circles). ac-field amplitude: 28 V/cm peak-to-peak. The solid lines joining the experimental points in all figures are only guides to the eyes.

For ac amplitudes varying from 5 to 30 V/cm, ε increases drastically and becomes frequency dependent, revealing the onset of domain dynamics as soon as the ac field exceeds 5 V/cm. This threshold value is extremely low compared to usual FE materials. This confirms the particularly soft FE behavior of RHSe. Above 50 V/cm, the permittivity reaches a nearly constant but frequency-dependent value. It should be noticed that no decrease of the permittivity is observed up to the highest ac field value of these measurements (400 V/cm).

The variations of the effective EO coefficient versus the amplitude of the ac field [Fig. 3(b)] reproduce the corresponding variations of the dielectric permittivity, at least in the range 20–350 V/cm where the EO measurements could be done (below 20 V/cm the EO signal was too weak to be measured within an acceptable accuracy). This correlation confirms that the contribution of domain dynamics to the EO coefficient is nearly proportional to the corresponding domain contribution to the dielectric susceptibility.

C. Frequency dispersion of the complex dielectric permittivity

The dispersions of ε' (real part) and ε'' (imaginary part) versus frequency were recorded under low ac amplitude (28 V/cm peak to peak), first near the coercive state ($E_{\rm dc}=-90$ V/cm) and then in the saturated state ($E_{\rm dc}=-450$ V/cm). The results are plotted in Fig. 4. The real part of the dielectric permittivity is nearly independent of the frequency when the sample is submitted to a strong dc field. This con-

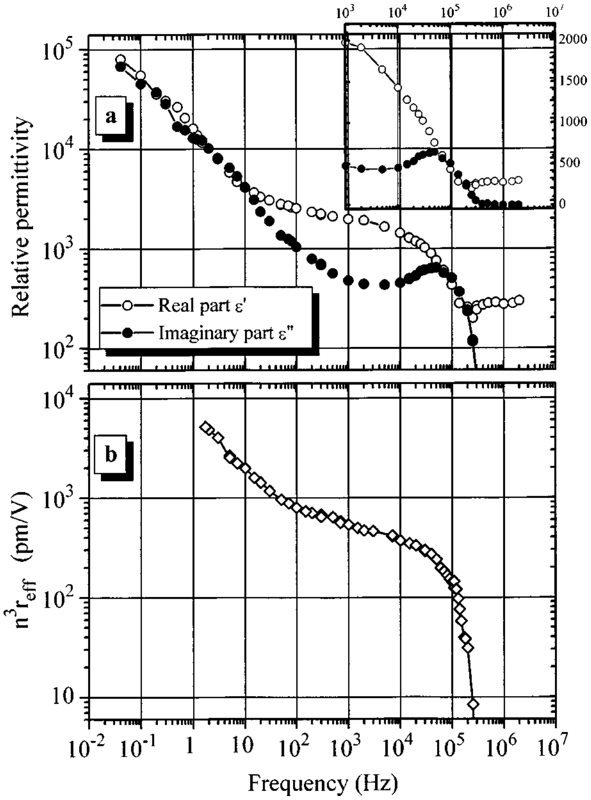

FIG. 5. Dispersions of the relative permittivity (a) and of the effective electro-optic coefficient (b). Measurements were made in the coercive state of the crystal under an ac-field amplitude of 125 V/cm peak to peak. The solid lines are only guides to the eyes.

firms that nearly all domains are reversed in the direction of the field and shows that no dispersion occurs below 100 kHz. Comparatively, the permittivity ε in the coercive state is much larger and frequency dependent. This is due to the contribution of domain reversal to the dielectric susceptibility. For instance, at 20 Hz this domain contribution is more than 15 times larger than the ionic, high-frequency, contribution. The low-frequency dispersion on the imaginary part of the permittivity (below 10 kHz) in the saturated state must be attributed to the ohmic contribution σ/ω, since the real part of the permittivity exhibits no dispersion in the corresponding frequency range. In the coercive state, this contribution is negligible because the dissipation due to domain dynamics remains largely dominant down to the low-frequency limit of this experiment (20 Hz).

The first piezoelectric resonances appear in the saturated state only, as a double peak around 160–300 kHz on the imaginary part of the permittivity (Fig. 4). These resonances are not visible in the coercive state: they are replaced by a broad Debye-like relaxation, which is more clearly evidenced under larger ac-field amplitudes [see the inset of Fig. 5(a)]. This relaxation can be attributed to the domain walls, which may impede the onset of a stationary acoustic wave along the length of the sample and thus hinders the corresponding piezoelectric resonances. However, another piezoelectric resonance at 800 kHz is visible in Fig. 4 in both the saturated state and the coercive state. This suggests that the corresponding acoustic wave vector is nearly parallel to the domain walls, so that this acoustic wave is not much affected by the presence of the domain structure.

D. Frequency dispersion of the EO coefficient

The dispersions $\varepsilon(f)$ and $n^3 r_{\mathrm{eff}}(f)$ reported in Fig. 5 were simultaneously recorded in the coercive state of the sample under an ac amplitude of 125 V/cm peak to peak. The frequency dispersions illustrate the close correlation between dielectric and electro-optic properties. The large EO effect related to domain dynamics remains appreciable up to the frequency of the first piezoelectric resonance. Above this frequency (160 kHz), the effective EO coefficient vanishes drastically and seems to reach a constant value of a few pm/V above 500 kHz. (This value could not be measured accurately because the high-frequency limit of our EO setup is about 500 kHz.) It thus seems that the direct Pockels effect due to the ionic contribution is rather weak in RHSe. However, this ionic contribution is expected to be practically flat versus frequency up to the microwave range, since the dielectric permittivity itself was shown to exhibit practically no dispersion up to 4 GHz.[11]

IV. INTERPRETATION

A. Domain contributions to the dielectric properties

All dielectric and EO responses observed in RHSe, i.e., with ac and dc fields and with frequency, can be attributed to both the specific domain structure and domain dynamics of this material. This domain structure appears under a polarizing microscope as layered strips, with domain thickness varying from 1 to 100 μm.[12] The so-called ''initial'' dielectric permittivity measured in the coercive state under small ac-field amplitudes is small and slightly frequency dependent (400 to 250). It can be mainly attributed to the ionic contribution, superimposed with a slight domain contribution probably due to domain-wall oscillations. This slight domain response remains linear with ac amplitude up to 5 V/cm. Under higher ac fields, the sharp increase of the susceptibility is probably due to another kind of domain motion, for which some domains begin to be switched as a whole by the electric field. This contribution increases with ac amplitude as more and more domains are set in motion. The very small threshold value of the ac field needed to start the domain reversal corroborates the particularly soft FE behavior of RHSe. This contribution of domain reversal leads to a plateaulike dependence versus ac amplitude in the range 30–400 V/cm. It suggests that nearly all the mobile domains are indeed set in motion by the ac field in this regime. For higher ac fields, however, one should expect a decrease of the permittivity, especially at very low frequency, if the ac amplitude would largely exceed the saturation value evidenced on the dc cycles (200 V/cm). This decrease of the permittivity is in fact not observed in Fig. 3(a), certainly because the lowest frequency of this experiment (5 Hz) is still far much higher than the recording frequency of the dc cycles (≈70 μHz).

It should be noticed that the frequency dispersion of both ε' and ε'' in the coercive state [Fig. 5(a)] does not look like a simple Debye relaxation. This is not surprising, since the domain structure is not fully regular, and domain dynamics proceeds from several kind of motions (small amplitude oscillations, large sideways motions, and domain-wall nucle-

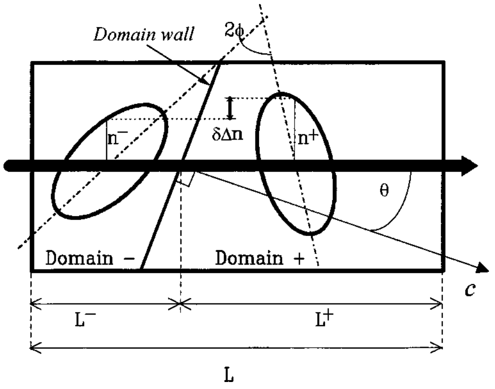

FIG. 6. Mutual tilt of the optical indicatrices in neighboring domains. When the propagation direction of the light is not perpendicular to domain walls, the birefringences Δn^+ and Δn^- in neighboring domains are different.

ations involved in switching processes). Most probably, several relaxation times are distributed in the relaxation phenomena.

The very soft FE properties of RHSe (low dc coercive field, low threshold ac amplitude for domain motion) seem somewhat exceptional compared to other ferroic materials [$(Gd_2(MoO_4)_3$ and $Bi_4Ti_3O_{12}$ crystals, $Pb(Zr,Ti)O_3$ ceramics]. This could be explained by the relatively moderate ferroelastic strains of the triclinic structure (α=90.7°, β=89.8°), probably associated with low elastic constants. To our knowledge, experimental data are not available for the elastic constants, but the relatively low frequencies of the piezoelectric resonances indicate that the sound velocities are small in this material. Consequently, the elastic energy involved in domain reversal is probably weak and thus allows easy motion of domain walls up to intermediate frequencies.

B. Domain contributions to the EO properties

The role played by domain dynamics in the large value of the electro-optic effect in RHSe originates from the tilting of the optical indicatrix caused by field-induced domain reversals. Since the ferroelectric domain structure of RHSe is also ferroelastic, the indicatrices in two neighboring domains are tilted each other by an angle.[9] Consequently, the birefringences Δn^+ and Δn^- in neighboring domains are not exactly equal, provided that the propagation direction of the light is not perpendicular to the domain walls (Fig. 6). We can thus express the birefringence along the light path length L as an average quantity through the polydomain sample

$$\Delta n=(L^+/L)\Delta n^+ + (L^-/L)\Delta n^-, \tag{1}$$

where L^+ and L^- are the light path lengths through domains "+" and "−" respectively.

The average ferroelectric polarization P of the sample can be expressed in the same way by

$$P=(L^+/L-L^-/L)P_S, \tag{2}$$

where P_S is the component of the spontaneous polarization parallel to domain walls, i.e., the $\mathbf{P_S}$ component that can be reversed by the electric field. The above equations take account of the contributions related to domain reversal only, the ionic contributions are assumed to be negligible in the present approach. Combining Eqs. (1) and (2), we can express the average birefringence under applied field as a function of the average ferroelectric polarization $P(E)$:

$$\Delta n(E)=\Delta n_0+\frac{1}{2}\,\delta\Delta n\frac{P(E)}{P_S}, \tag{3}$$

where $\Delta n_0=(\Delta n^+ + \Delta n^-)/2$ is the natural birefringence of the sample in the coercive state ($P=0$), and $\delta\Delta n=(\Delta n^+ - \Delta n^-)$ is the difference between the birefringences of two neighboring domains. This term $\delta\Delta n$ depends on the angle of incidence θ of the light beam with respect to the normal of domain walls. Obviously, the birefringence reaches, in the fully saturated states ($P=\pm P_S$), the corresponding saturation value $\Delta n=\Delta n^+$ or Δn^-. Here we have also neglected the influence of the FEL strains, which should lead in Eq. (3) to corrective terms of the second and third orders in powers of $P(E)/P_S$. Obviously, these terms are small compared with the first-order term provided that the angle of incidence θ of the light beam with respect to the normal of the domain walls is much larger than the angles of the FEL shear strains, which are equal to +0.6° and −0.2° in the planes (100) and (010), respectively.[5]

Taking the derivative of Eq. (3) versus the electric field E, we obtain

$$\frac{d\Delta n}{dE}=\frac{\delta\Delta n}{2P_S}\frac{dP}{dE}. \tag{4}$$

By definition, the derivative of the field-induced birefringence yields the effective EO coefficient, while the derivative of the ferroelectric polarization gives the dielectric susceptibility (multiplied by the permittivity of vacuum ε_0). We thus obtain a direct relationship between the domain contribution to the EO coefficient $n^3 r_{\mathrm{eff},d}$ and the domain contribution to the dielectric susceptibility χ_d:

$$\frac{1}{2}n^3 r_{\mathrm{eff},d}(E_{\mathrm{ac}},\omega)=\frac{\delta\Delta n}{2P_S}\varepsilon_0\chi_d(E_{\mathrm{ac}},\omega). \tag{5}$$

Since the domain-related susceptibility χ_d strongly depends on the amplitude and on the frequency of the ac field, the EO effect should follow similar nonlinearity and similar frequency dispersion as χ_d, as far as domain dynamics is concerned in the dielectric response—that is, in the low-frequency range up to the vicinity of the piezoelectric resonances. The tight correlation between the dielectric response and the EO response is clearly evidenced in Fig. 7: the curve $n^3 r_{\mathrm{eff}}$ versus ε exhibits a constant slope in the frequency range 1 Hz–10 kHz, revealing the preponderance of domain dynamics. In the range 10–100 kHz, a slight decrease of the slope is observed, since the contribution of domain dynamics to the EO effect begins to decrease. At higher frequencies, the domain contribution vanishes drastically [see Fig. 5(b)] Eq. (5) is no longer relevant to describe the relationship between dielectric and electro-optic properties but has to be replaced by more usual models.[2,13]

Finally, it is worth paying attention to the coefficients of the Pockels tensor that should be involved in the domain contribution. In the case of RHSe the indirect Pockels effect due to domain dynamics must be analyzed in the monoclinic

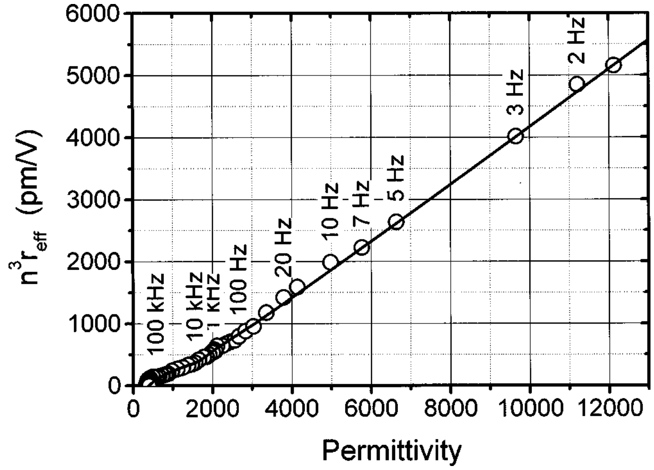

FIG. 7. Correlated dispersions of the dielectric permittivity and the effective electro-optic coefficient. The experimental points (circles) are directly deduced from the measurement reported in Fig. 5. The solid line is a multiple Debye fit involving the same set of relaxation times for both ε and n^3r_{eff}.

supergroup ($P2$) of the paraelectric phase. Since the electric field has to be directed close to the ferroelectric b axis to induce efficient domain reversals, the EO coefficients involved in the domain-related EO effect have to belong to the second column of the Pockels tensor. In this column, two coefficients are allowed by the symmetry of the supergroup: r_{42} and r_{52}. These two coefficients describe the field-induced tilt motions of the optical indicatrix, around the x_1 axis and x_2 axis, respectively, which are defined as the optical neutral lines in the (a,b) plane of domain walls.[12] We have recently measured[12] the natural tilt angles of the optical indicatrix with respect to domain walls (Fig. 6) in polydomain RHSe crystals: at 633 nm the tilt angle ϕ_1 around the x_1 axis (2.2°) is larger than the tilt angle ϕ_2 around the x_2 axis (0.6°). Additionally, it was also found[12] that the corresponding birefringence Δn_1 (0.0560 at 633 nm) is much larger than Δn_2 (0.0139 at 633 nm). Since the indirect Pockels coefficients r_{42} and r_{52} related to domain dynamics should be proportional to the corresponding products $\Delta n_i \times \sin 2\phi_i$, it can be expected that the largest Pockels coefficient of RHSe in the polydomain state should be r_{42}. This prediction has now to be confirmed by further electro-optic experiments on RHSe samples cut at 45° from the x_2 (resp. x_1) axis towards the c axis and allowing direct measurements of the coefficients r_{42} and r_{52}.

V. CONCLUSION

We have shown that the dielectric and electro-optic properties in the crystal of RHSe are tightly correlated to each other and closely linked to the domain dynamics. A model that accounts for the difference in the birefringence between neighboring domains and the reversal of these domains yields the relationship between the permittivity and the EO coefficient and is able to explain the huge values of the EO properties recorded in this material. In this way, we have pointed out a particular EO mechanism related to the domain dynamics that could lead to an enhancement of the EO properties in any materials that possess both ferroelectric and ferroelastic properties.

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