

High microwave frequency EPR study of the axial $\text{Fe}^{3+}\text{-V}_\text{O}$ centers in the quantum paraelectrics SrTiO_3 and KTaO_3

Ikram Zdeg , Oleksii Laguta ,* and Petr Neugebauer [†]*Central European Institute of Technology, Brno University of Technology, 61200 Brno, Czech Republic*Valentyn Laguta *Institute of Physics, Czech Academy of Sciences, 18200 Prague, Czech Republic*

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Oxygen vacancies are the most common defects in oxide materials. They frequently couple with transition-metal ions to form charge-compensated paramagnetic Me-V_O pair centers, which exhibit large zero-field splitting of $2.6\text{--}6\text{ cm}^{-1}$. Such splitting requires the use of high microwave frequencies in electron paramagnetic resonance (EPR) spectroscopy. Using high-frequency EPR (up to 210 GHz), we investigated $\text{Fe}^{3+}\text{-V}_\text{O}$ centers in the quantum paraelectrics SrTiO_3 and KTaO_3 over the temperature range 5–296 K. These materials have attracted significant interest due to their unique dielectric properties and recently discovered spin-electric phenomena, including two-dimensional superconductivity and the anomalous Rashba effect in spin-charge conversion. To probe spin-electric coupling, we also performed pulse EPR measurements at 9.79 GHz under applied electric fields. Our results show that the axial crystal-field parameter increases nearly linearly from 13.1 GHz in KTaO_3 and 13.55 GHz in SrTiO_3 at room temperature to $\simeq 14.6$ GHz at 20 K, below which it saturates. This temperature dependence is attributed to anomalous displacements of four lateral oxygen ions relative to Fe^{3+} , likely governed by the soft optical mode. Application of electric fields up to 30 kV/cm revealed not only splitting of the EPR lines but also redistribution of domain populations, observable at fields as low as 10 kV/cm in the nonpolar tetragonal phase of SrTiO_3 .

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I. INTRODUCTION

Quantum paraelectrics such as strontium titanate (SrTiO_3) and potassium tantalate (KTaO_3) exhibit dielectric properties close to a ferroelectric phase transition but remain paraelectric even at very low temperatures, as quantum fluctuations suppress long-range order. These systems display quantum critical behavior, making them important platforms for studying quantum phase transitions, dielectric responses, and emergent electronic phenomena [1–3].

Recently, attention has shifted toward KTaO_3 , which shares the dielectric properties of SrTiO_3 but benefits from the much stronger spin-orbit coupling of the heavy $5d$ Ta ion compared to the $3d$ Ti ion [4]. The unique dielectric properties of SrTiO_3 and KTaO_3 can be utilized in the nonvolatile electric control of spin-charge conversion in a Rashba system that has been recently demonstrated for both these materials [5,6], in quantum information processing for hosting of spin qubits [7] and quantum sensing [8]. Moreover, as we have recently

demonstrated [9], in quantum paraelectrics the power of microwave pulses required for coherent spin manipulation on the nanosecond timescale can be reduced to the milliwatt level, in contrast to the tens or hundreds of watts typically required in conventional materials. This greatly relaxes the need for expensive high-power microwave sources.

A promising strategy for spin control in these materials is the use of dipole paramagnetic centers, which possess both an electric dipole and an electron spin. Such centers allow electrical tuning of the spin Hamiltonian of S -state ions ($L = 0$, long relaxation times), manipulation of spin superposition phases, and, in principle, driving of spin transitions with resonant microwave electric fields. In cubic SrTiO_3 and KTaO_3 lattices, an important example is the $\text{Fe}^{3+}\text{-V}_\text{O}$ center, formed by a $3d^5$ Fe^{3+} ion ($S = 5/2$, $L = 0$) adjacent to an oxygen vacancy. This center has been extensively studied in SrTiO_3 , where it is highly sensitive to the cubic-to-tetragonal structural phase transition at 105 K [10–12]. Its presence in KTaO_3 was reported more recently [13]. The $\text{Fe}^{3+}\text{-V}_\text{O}$ center exhibits a strong linear electric-field effect on Fe^{3+} spins, even for the central $-1/2 \longleftrightarrow +1/2$ transition, due to rotation of the magnetic axes in an electric field [14].

Previous studies, however, have been largely limited to low microwave frequencies (9–20 GHz), where only the central transition can be observed. The large zero-field splitting of $2.6\text{--}5.4\text{ cm}^{-1}$ prevents direct access to the full set of crystal-field parameters. Transitions between the two lowest Kramers doublets were later detected in SrTiO_3 at 35–75 GHz, but only

*Contact author: oleksii.laguta@ceitec.vutbr.cz[†]Contact author: petr.neugebauer@ceitec.vutbr.cz

at room temperature [15,16]. As a result, spin Hamiltonian parameters have so far been determined only approximately in SrTiO₃, while studies in KTaO₃ remain scarce. Furthermore, the splitting of spin levels is strongly temperature dependent, complicating accurate determination of the spin Hamiltonian parameters—particularly in SrTiO₃, where the structural transition at 105 K adds further complexity.

In this work, we report a high-frequency (up to 210 GHz) EPR study of Fe³⁺-V_O centers in KTaO₃ and SrTiO₃. Fully resolved Fe³⁺-V_O spectra were obtained in single crystals, enabling precise determination of crystal-field parameters and their temperature dependence in the range 5–296 K. We also discuss the role of the soft optical mode, whose frequency is comparable to the applied microwave frequencies, in shaping the EPR spectra and crystal-field parameters. In addition, pulse EPR measurements were performed under applied electric fields up to 30 kV/cm. Finally, it should be emphasized that high-frequency EPR is particularly well suited for materials with large dielectric permittivity, as the absence of a resonant cavity minimizes perturbations of the microwave fields by the sample.

II. EXPERIMENTAL METHODS

EPR measurements were performed on single-crystal samples with typical rectangular dimensions of 3 × 2 × 0.3 mm³ with edges aligned along the cubic (100) axes. A commercial SrTiO₃ crystal (MSE Supplies, USA) containing 0.05 wt% Fe was used, providing sufficient concentration for EPR measurements. The KTaO₃ crystal was grown by the self-flux solution method [17] and doped with Fe to a concentration of ≈0.1 wt%.

For measurements under applied electric fields, electrodes were deposited on the largest crystal faces using silver paste (SPI-paint). The samples were electrically insulated with picein compound, enabling the application of electric fields up to 30 kV/cm.

High-frequency EPR measurements (150–210 GHz) were carried out using a home-built induction mode spectrometer operating in the 90–1100 GHz range, equipped with a 16 T superconducting magnet (Cryogenic Ltd.), a quasi-optical bridge, and a superheterodyne receiver [18–20]. EPR measurements with applied electric fields were performed using a Bruker E580 pulse spectrometer, equipped with an MD5 dielectric ring resonator at 9.79 GHz, in the temperature range 5–14 K. Both spectrometers were fitted with liquid helium cryostats, enabling measurements between 2.5 and 296 K.

III. RESULTS AND DISCUSSION

A. Fe³⁺-V_O spectra in KTaO₃

EPR spectra of the Fe³⁺-V_O center were measured in KTaO₃ at 150 and 210 GHz in the temperature range 5–296 K. Representative spectra recorded for magnetic field **B**||[100] at 296 and 125 K are shown in Figs. 1 and 2.

Because the oxygen vacancy in the Fe³⁺-V_O center can occupy any of the six ⟨100⟩ directions with equal probability, the spectra contain contributions from six centers. However, due to cubic symmetry of lattice, these reduce to three magnetically nonequivalent centers. For **B**||[100], one center has

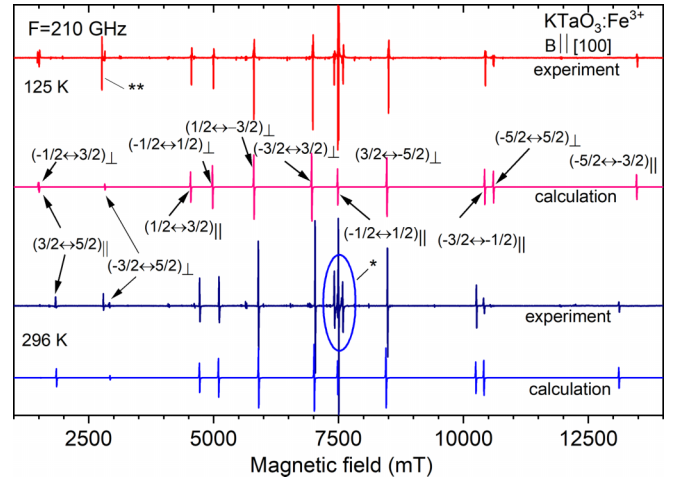


FIG. 1. EPR spectrum of the Fe³⁺-V_O center in KTaO₃ measured at 210 GHz and the temperatures 296 and 125 K together with the simulated spectra. $M_{S-1} \longleftrightarrow M_S$ spin transitions are labeled according to their $M_S = \langle S_Z \rangle$ projections when a magnetic field is applied along the [100] axis. The group of lines around 7500 mT belongs to the Fe³⁺ cubic symmetry center. The single spectral line denoted by the symbol “*” is not identified. The “||” and “⊥” symbols denote the magnetic field direction along or perpendicular to the center’s axial axis.

its magnetic axis parallel to the field, while the other two are perpendicular. The corresponding spectral lines are labeled “||” and “⊥” in Figs. 1 and 2. All five spin transitions allowed for $S = 5/2$ are clearly resolved in the spectra. The spectra were simulated using the EasySpin toolbox [21] with the following spin Hamiltonian:

$$\mathbf{H} = \beta \mathbf{SgB} + B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4, \quad (1)$$

where B_l^m are crystal-field parameters and O_l^m electron spin polynomials (Stevens spin operators) [22]. The fitted spin

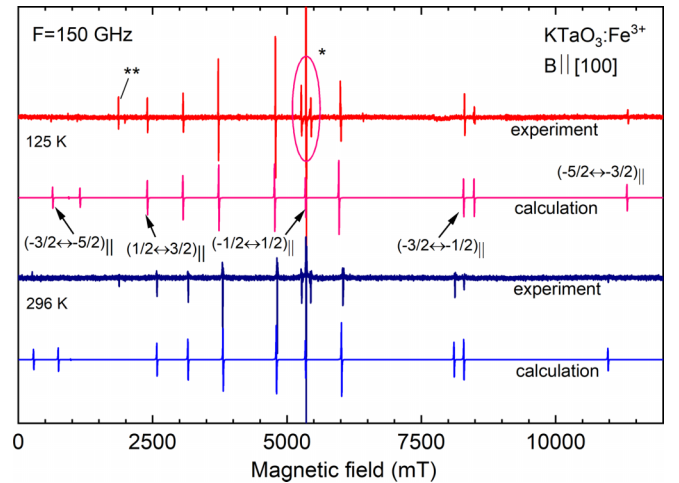


FIG. 2. EPR spectrum of the Fe³⁺-V_O center in KTaO₃ measured at 150 GHz, and the temperature 296 and 125 K together with the simulated spectra. Spin transitions are labeled for the spectrum measured at 125 K and only for the center with **B**||[100]. The “*” and “***” symbols denote the same resonances as in Fig. 1.

TABLE I. Spin Hamiltonian parameters of $\text{Fe}^{3+}\text{-V}_O$ centers in KTaO_3 and SrTiO_3 . Crystal-field parameters are given in MHz.

Material	T(K)	g factor	B_2^0	B_2^2	B_4^0	B_4^4	Frequency (GHz)	Reference
KTaO_3	296	2.005	13100	0	3.3	16	150–210	This work
	125	2.005	13930	0	3.3	16	150–210	This work
	5	2.005	14500	0	3.3	16	150–210	This work
SrTiO_3	290	$g_{\parallel} = 2.0037$ $g_{\perp} = 2.006$	13550	0	1.0	5.0	150–210	This work
	260	$g_{\parallel} = 2.0037$ $g_{\perp} = 2.006$	13650	0	1.0	5.0	150–210	This work
	47	$g_{\parallel} = 2.0037$ $g_{\perp} = 2.006$	14550		1.0	5.0	150–210	This work
	14	$g_{\parallel} = 2.0037$ $g_{\perp} = 2.006$	14650	90	1.0	5.0	150–210	This work
	296		13650 ^a	0	3.3 ^a	16.5 ^a	35,70	[15,16]
	78	$g_{\parallel} = 2.0037$	13650	38.8			K band	[10]

^a B_l^m parameters are recalculated from zero-field splitting data of Ref. [15].

Hamiltonian parameters for three temperatures are presented in Table I. The corresponding energy-level diagram of the $\text{Fe}^{3+}\text{-V}_O$ center with indicated spin transitions is shown in Fig. S1 of the Supplemental Material [23]. Note that the spin transition probabilities for the three centers are not equal. The center with the axial axis pointed along the oscillating magnetic field direction has a much lower probability of spin transitions compared to two other centers where the oscillating field is perpendicular to the center's axis. The mixing of spin states for the perpendicular direction of the magnetic field also changes the transition probabilities.

The axial parameter B_2^0 exhibits a pronounced increase on cooling, despite the lattice constant being only weakly temperature dependent [24]. This suggests a strong coupling of the $\text{Fe}^{3+}\text{-V}_O$ center to lattice vibrations associated with the soft optical mode (discussed below).

Besides the $\text{Fe}^{3+}\text{-V}_O$ center, spectral lines originated from the cubic-symmetry Fe^{3+} center (without a nearby oxygen vacancy) are observed near $g = 2.00$, denoted by circles in Figs. 1 and 2 [13]. The unidentified line marked “*” most likely arises from a second Fe^{3+} axial center characterized by $B_2^0 \simeq 44.6$ GHz [25].

B. $\text{Fe}^{3+}\text{-V}_O$ spectra in SrTiO_3

Fe^{3+} EPR spectrum in SrTiO_3 was also measured at microwave frequencies 150 and 209.96 GHz in the temperature range 25–296 K. Representative spectra at 150 GHz recorded at 260 K (above the cubic-to-tetragonal transition) and at 47 K (in the tetragonal phase) are shown in Fig. 3, while Fig. 4 presents the spectrum measured at 209.96 GHz and 116 K. The extracted spin Hamiltonian parameters for both phases are listed in Table I.

Below the structural phase transition at 105 K, the analysis of $\text{Fe}^{3+}\text{-V}_O$ spectrum requires addition of a rhombic term $B_2^2 O_2^2$ to the Hamiltonian (1), consistent with distortions induced by the rotation of the oxygen octahedra [10]. Notably, the intensity of the $\text{Fe}^{3+}\text{-V}_O$ spectrum in SrTiO_3 decreases sharply below 25 K at high frequencies (150–210 GHz), rendering the signal unobservable at lower temperatures. This behavior is attributed to the long spin-lattice relaxation time and

possible microwave absorption by the crystal, since the excitation frequency approaches that of the electric-active soft optical mode. Additionally, this absorption is further promoted by the fact that the crystal interacts with both magnetic and electric components of the microwave as no resonator is used in the high-frequency spectrometer.

C. Temperature dependence of the crystal-field parameters of the $\text{Fe}^{3+}\text{-V}_O$ centers

Figure 5 shows the temperature dependence of the axial crystal field parameter B_2^0 for $\text{Fe}^{3+}\text{-V}_O$ centers in KTaO_3 and SrTiO_3 extracted from spectra measured at 150 and 210 GHz. EPR intensity in SrTiO_3 sharply decreases to zero below approximately 25 K as noted above. Therefore, complementary pulse EPR measurements at 9.79 GHz were used to determine B_2^0 from the $-3/2 \longleftrightarrow +3/2$ transition at 880 mT at the

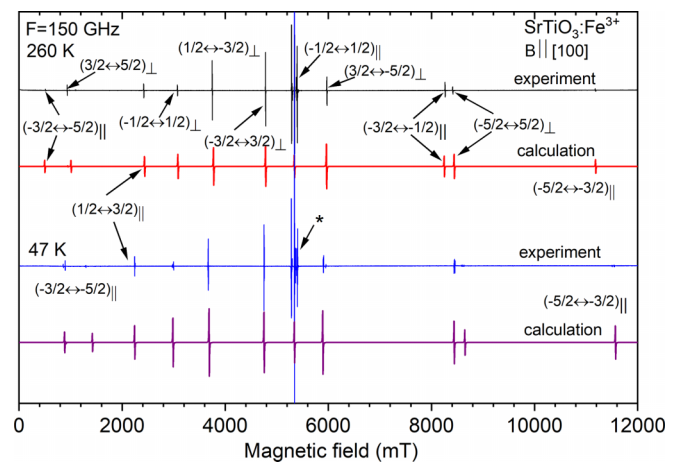


FIG. 3. EPR spectrum of $\text{Fe}^{3+}\text{-V}_O$ center in SrTiO_3 measured at 150 GHz and the temperature 260 and 47 K including the simulated spectra with indicated spin transitions. The symbol “*” denotes spectral lines from cubic symmetry Fe^{3+} and Cr^{3+} centers which are superimposed on the $-1/2 \longleftrightarrow 1/2$ central transition of $\text{Fe}^{3+}\text{-V}_O$ center.

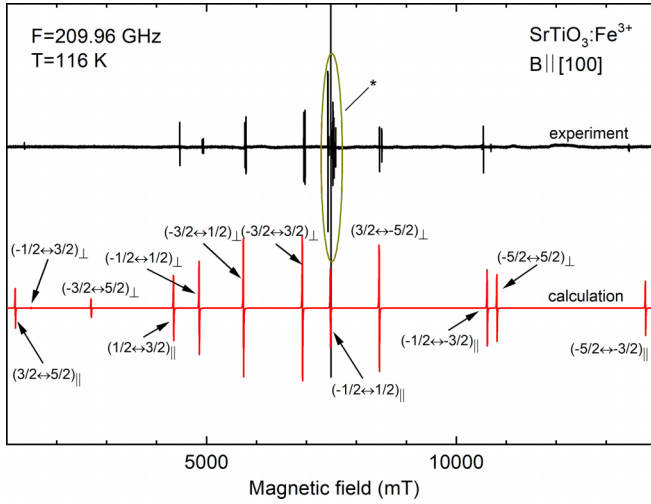


FIG. 4. EPR spectrum of $\text{Fe}^{3+}\text{-V}_\text{O}$ center in SrTiO_3 measured at 209.96 GHz and the temperature 116 K including the simulated spectrum with indicated spin transitions. The symbol “*” denotes spectral lines from cubic symmetry Fe^{3+} and Cr^{3+} centers.

lowest temperatures. These results are included in Fig. 5 (blue squares).

Our data reveals a nearly linear increase in the crystal field parameter upon cooling from room temperature to about 25 K, below which the values level out. Overall, the axial crystal-field parameter increases by 13% in KTaO_3 and $\simeq 8\%$ in SrTiO_3 between room temperature and the low-temperature plateau. In both materials, the crystal lattice constant decreases by only 0.15–0.2% on temperature lowering from 300 K to 5 K [24,26], and it is too small to produce such large variations of B_2^0 . Therefore, we must consider local structural distortions of the $\text{Fe}^{3+}\text{-V}_\text{O}$ center and the dependence of the Fe^{3+} ion crystal field parameters on its immediate environment.

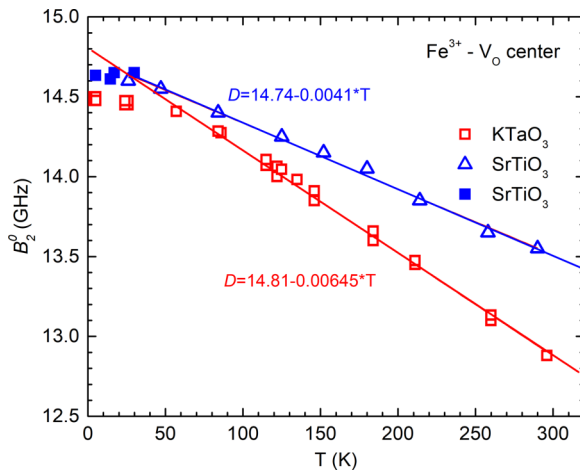


FIG. 5. Temperature dependence of the B_2^0 crystal-field parameter for $\text{Fe}^{3+}\text{-V}_\text{O}$ center in KTaO_3 and SrTiO_3 . For SrTiO_3 blue open triangles show data obtained at the frequency 150 GHz and blue filled squares show data obtained at the frequency 9.790 GHz using pulse EPR technique. Solid lines are linear fit.

The microscopic structure of the $\text{Fe}^{3+}\text{-V}_\text{O}$ center in oxide perovskite compounds was analyzed by Siegel and Muller [27] using the Newman superposition model. They found that the Fe^{3+} ion shifts by $\simeq 0.2$ Å toward the vacancy, while the four lateral oxygen ions move oppositely by 2–4%. This displacement of 0.04–0.08 Å is consistent with the reduction of the Fe^{3+} ionic radius when its coordination number decreases from six to five [28]. Subsequent *ab initio* [29] and atomistic [30] calculations confirmed this model. We reproduced the calculation of the axial-field parameter B_2^0 for the $\text{Fe}^{3+}\text{-V}_\text{O}$ center in both SrTiO_3 and KTaO_3 with parameters of the Newman model taken from Ref. [27] with the following expression:

$$B_2^0 = \frac{1}{3} b_2(R_0) \left[\sum_i \left(\frac{R_0}{R_i} \right)^{t_2} K_2^0(\theta_i, \phi_i) \right], \quad (2)$$

where for the case of axial symmetry $K_2^0 = \frac{1}{2}(3 \cos^2 \theta - 1)$. The reference distance R_0 is 1.952 and 1.994 Å for SrTiO_3 and KTaO_3 , respectively, and R_i is the radial metal-ligand distance. The angle θ defines the angle between axial axis and direction to the ligand ion. Parameter b_2 was taken from Ref. [27]: $-0.74(11)$ and $-0.63(8)$ cm^{-1} for SrTiO_3 and KTaO_3 , respectively, and the t_2 parameter was 8.

The results are shown in Fig. 6. Here, parameter c defines the displacements of the four planar oxygen ions as $d = ca/2$, where a is the lattice constant. Horizontal lines indicate the maximal and minimal experimental values of B_2^0 constant according to the data in Fig. 5. The calculated curves exhibit broad maximum for Fe^{3+} displacement of 0.2–0.3 Å but the resulting B_2^0 values remain significantly below the experimental ones if the planar oxygens are not shifted ($c = 0$). Agreement with the room-temperature experimental B_2^0 values requires planar oxygen contractions of $c = 0.021$ (SrTiO_3) and $c = 0.026$ (KTaO_3), corresponding to their ionic displacements of $d = 0.04$ and $d = 0.052$ Å, respectively. Because B_2^0 depends only weakly on the Fe^{3+} ion shift, the temperature dependence of B_2^0 is dominated by the displacement of the four planar oxygens. At 5–20 K, the predicted displacements of planar oxygen ions reach 0.056 Å for SrTiO_3 and 0.076 Å for KTaO_3 , values about 16 times larger than expected from thermal contraction of the lattice alone [24,26].

This anomalously large displacement is most likely associated with the softening of the polar-active optical mode, whose frequency decreases to $\simeq 10$ cm^{-1} in SrTiO_3 and $\simeq 15$ cm^{-1} in KTaO_3 for $T < 10$ K [31,32]. Quantum fluctuations at these temperatures further contribute to the saturation of both the soft-mode frequency and the crystal-field parameter. This interpretation is consistent with similar temperature-dependent changes of crystal-field parameters observed in other paramagnetic centers, including the cubic-symmetry Fe^{3+} center in both materials and Mn^{2+} centers in KTaO_3 [13,33].

D. Effect of electric field on $\text{Fe}^{3+}\text{-V}_\text{O}$ center

Application of an electric field to the crystals is an elegant way of testing the dominant role of the four planar oxygen ions. For this, we have chosen SrTiO_3 where spectral lines are narrow and sufficiently intense to measure the nominally

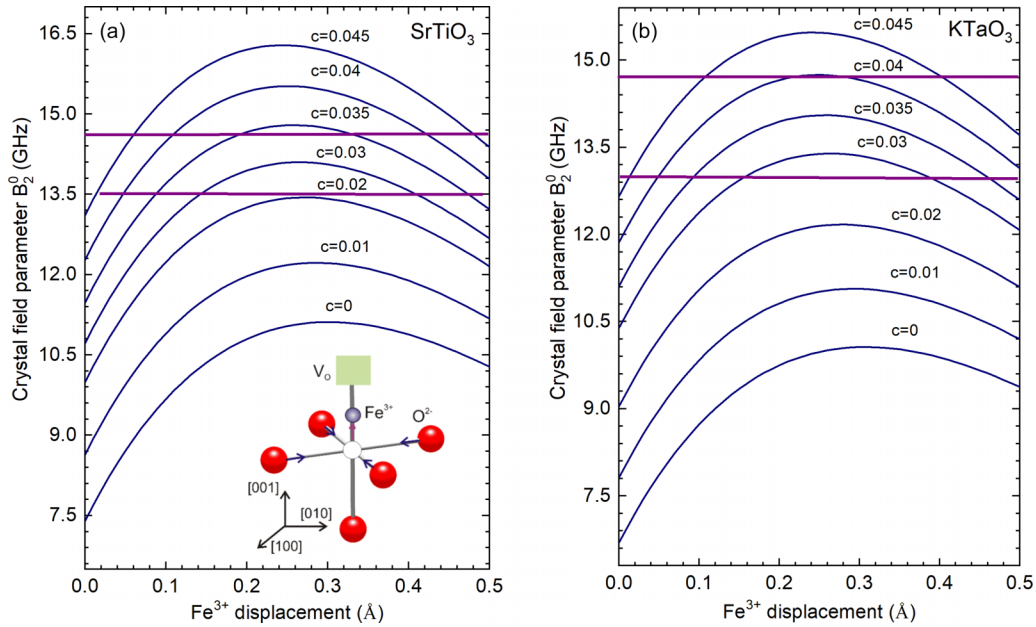


FIG. 6. Dependence of the axial crystal-field parameter B_2^0 on Fe^{3+} off-center shift toward the oxygen vacancy with different contractions (parameter c) of the four planar oxygen ions in (a) SrTiO_3 and (b) KTaO_3 . The horizontal lines define regions of experimental B_2^0 constant value and its change with temperature. The inset in panel (a) illustrates the model of the Fe^{3+} - V_O center, where arrows show displacements of oxygen and Fe ions.

forbidden $-3/2 \longleftrightarrow +3/2$ transition by X-band (≈ 9.8 GHz) pulse EPR at temperatures 10–15 K. In the case of KTaO_3 though, the spectral width of this transition is too large and, consequently, EPR intensity is too weak for doing reliable experiments with the electric field. Figure 7 shows the results obtained at 14 K under applied fields up to 30 kV/cm, together with the cubic-phase spectrum recorded at 295 K for comparison. The inset illustrates the relative orientations of the electric and magnetic fields with respect to the Fe^{3+} - V_O center axes. Notably, at these microwave frequencies the $-3/2 \longleftrightarrow +3/2$ transition is observable only for the magnetic field direction nearly perpendicular to the center axis due to the mixing of states. This is illustrated by simulated energy diagrams and corresponding spectrum presented in Fig. S2 of the Supplemental Material for cubic phase [23].

In the tetragonal phase, the multidomain structure causes splitting of the single spectral line appearing in the cubic phase into multiple (up to five) components. The tetragonality in SrTiO_3 originates from rotation of oxygen octahedrons around $\langle 100 \rangle$ pseudocubic directions and the rotation angle ϕ is the phase order parameter. At the magnetic field direction along $\langle 100 \rangle$ pseudocubic directions, some resonances coincide and only three components are resolved as shown in Fig. 7 (spectrum at zero electric field, $E = 0$). Waldkirch *et al.* conducted a detailed analysis of the central transition $-1/2 \longleftrightarrow +1/2$ behavior in the tetragonal phase [10]. Following their results, we apply this analysis to the high-field $-3/2 \longleftrightarrow +3/2$ transition which is much more sensitive to changes of the crystal-field splitting parameters. In particular, the central component (2) belongs to the domain with the tetragonal distortion along [100] and [010] pseudocubic directions, i.e., when tetragonality appears along axial axis of Fe^{3+} - V_O center, and thus, it does not change the center's

symmetry. Moreover, B_2^0 constant does not change because of very weak tetragonal distortion of the crystal in the tetragonal phase, $c/a \approx 3.7 \times 10^{-4}$ at 20 K [26]. However, when the tetragonality arises perpendicularly to the axial axis, it lowers the symmetry of Fe^{3+} - V_O center to the orthorhombic one and causes splitting of the spectral line. These "orthorhombic" lines are labeled by (1) and (3) in Fig. 7. They are double degenerated and correspond to centers that are related to one another by the inversion operation and the (001) mirror plane. The simulated spectrum of the orthorhombic symmetry centers assuming equal population of tetragonal domains is shown in Fig. 7 by dotted lines and the corresponding spin Hamiltonian parameters are listed in Table I. In particular, the rhombic crystal-field parameter B_2^2 is 90 MHz. The degeneracy of spin transitions is removed by electric field applied along the [001] direction, i.e., perpendicularly to center's axis as illustrated in Fig. 7 since centers have no inversion symmetry. Remarkably, even at 10 kV/cm the intensity of component (1) decreases strongly, indicating redistribution of domain populations in the electric field. It is somewhat surprising since the tetragonal phase of SrTiO_3 is nonpolar and therefore domains must be nonelectrically active.

The observed behavior indicates that the electric field primarily displaces O^{2-} and Fe^{3+} ions in opposite directions, altering the Fe–O distances and, consequently, modifying the crystal-field parameters. However, the situation is more complex: at low temperatures the SrTiO_3 lattice is highly polarizable, and an applied field not only perturbs the Fe^{3+} - V_O center locally but also induces polarization of the surrounding lattice. Moreover, even moderate electric fields significantly affect the dielectric permittivity [34–37]. As a result, the effect of electric fields on the Fe^{3+} - V_O center in tetragonal SrTiO_3 is multifaceted and requires further detailed study. Nevertheless,

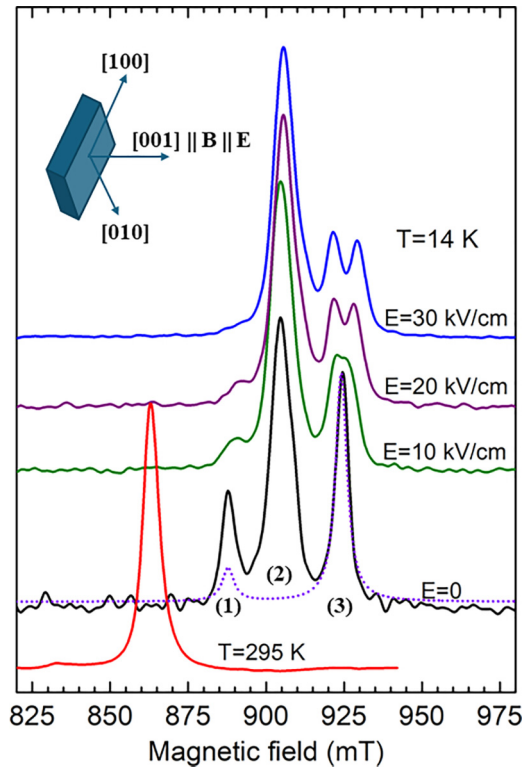


FIG. 7. $-3/2 \longleftrightarrow +3/2$ transition spectral line of $\text{Fe}^{3+}\text{-V}_\text{O}$ center in the tetragonal phase of SrTiO_3 ($T = 14$ K) at zero electric field and applied electric field of 10, 20, and 30 kV/cm normalized to the intensity of the central line (2). Both magnetic and electric fields are parallel to each other and define center with axial axis along the $[001]$ direction that is normal to the sample plane as shown in the inset. Other two centers have axial axes in the plane of sample. Numbers (1)–(3) for the low-temperature spectra numerate split components of orthorhombic symmetry centers and the dotted line shows calculated spectrum for these centers. For comparison, bottom spectrum in red is measured in the cubic phase at 295 K.

the observed line splittings and domain redistribution demonstrate the high mobility of the $\text{Fe}^{3+}\text{-V}_\text{O}$ local environment under relatively modest electric fields.

IV. CONCLUSIONS

We have performed high-frequency EPR measurements of $\text{Fe}^{3+}\text{-V}_\text{O}$ centers in the quantum paraelectrics KTaO_3 and SrTiO_3 at microwave frequencies up to 210 GHz and temperatures down to 5 K. Owing to the presence of oxygen

vacancies, these centers exhibit very large zero-field splitting ($2.6\text{--}6\text{ cm}^{-1}$). High-frequency EPR enabled full resolution of all spin transitions of the $S = 5/2$ system, allowing more accurate determination of spin Hamiltonian parameters than in previous low-frequency studies.

The axial crystal-field parameter B_2^0 was found to increase linearly with cooling, from 13.1 GHz in KTaO_3 and 13.55 GHz in SrTiO_3 at room temperature to a common value of $\simeq 14.6$ GHz at 20 K, below which it saturates. This behavior is explained by anomalous displacements of the four lateral oxygen ions toward the Fe^{3+} paramagnetic ion. Calculations based on the Newman superposition model yield predicted oxygen displacements of $0.04\text{--}0.052\text{ \AA}$ at room temperature and $0.056\text{--}0.076\text{ \AA}$ at 5–20 K, values 10–16 times larger than those expected from thermal contraction alone. We attribute this effect to the “soft” character and high polarizability of quantum paraelectric lattices that are determined by the presence of optical soft mode.

In SrTiO_3 , additional pulse EPR measurements at $\simeq 9.8$ GHz under applied electric fields up to 30 kV/cm revealed not only splitting of EPR lines but also redistribution of domain populations, observable already at 10 kV/cm in the primarily nonpolar tetragonal phase. This suggests that electric fields effectively polarizing the lattice suppress octahedral rotations along certain directions while favoring others. Electrostriction can also affect the domain structure. However, the actual mechanism of this phenomenon requires a separate detailed investigation.

Applied by us, high-frequency EPR technique as well as pulse EPR are very suitable for study of materials with high dielectric permittivity. High-frequency EPR spectrometer does not use microwave resonator. Thus, microwave fields are not much perturbed by high-permittivity samples. In pulse EPR measurements, the resonator is tuned to the smallest Q factor that practically completely diminishes influence of the sample’s high permittivity on the resonator tuning. All these advantages are especially useful when the dielectric permittivity in material changes very sharply with temperature.

ACKNOWLEDGMENT

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DATA AVAILABILITY

The data that support the findings of this article are openly available [38], embargo periods may apply.

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