

Unconventional Magnetostructural Transition in CoCr_2O_4 at High Magnetic Fields

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The magnetic-field and temperature dependencies of the ultrasound propagation and magnetization of single-crystalline CoCr_2O_4 have been studied in static and pulsed magnetic fields up to 14 and 62 T, respectively. Distinct anomalies with significant changes in the sound velocity and attenuation are found in this spinel compound at the onset of long-range incommensurate-spiral-spin order at $T_s = 27$ K and at the transition from the incommensurate to the commensurate states at $T_l = 14$ K, evidencing strong spin-lattice coupling. While the magnetization evolves gradually with the field, steplike increments in the ultrasound clearly signal a transition into a new magnetostructural state between 6.2 and 16.5 K and at high magnetic fields. We argue that this is a high-symmetry phase with only the longitudinal component of the magnetization being ordered, while the transverse helical component remains disordered. This phase is metastable in an extended $H - T$ phase space.

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Multiferroic materials, which exhibit concomitant magnetic and ferroelectric order, are of great current interest, both from a fundamental as well as an application-oriented view. They challenge our understanding of ordering phenomena but in addition provide new functionalities in spintronics since these dielectric and magnetic polarizations can be tuned by either external magnetic or electric fields [1–4]. Among multiferroics, magnetic AB_2X_4 compounds with a spinel structure attracted considerable interest, revealing colossal magnetocapacitance and spontaneous dielectric polarization in the magnetically ordered state [5–9]. The appearance of dielectric polarization is associated either with a noncollinear arrangement of spins, with charge order, or with magnetic ions moving off center from their symmetric site positions in the lattice due to strong magnetoelastic effects.

Significant spin-lattice coupling and magnetic frustration are important features of spinels, specifically, of chromium oxides and chalcogenides. Despite quenched orbital moments, these compounds reveal structural instabilities which are governed explicitly by the ordering of spins, e.g., giant magnetostriction, negative thermal expansion [10], and spin-Jahn-Teller instabilities [11–15]. In Cr spinels with only one magnetic sublattice, where the Cr^{3+} ions are located solely on the pyrochlore lattice of the B sites, it is well known that the oxides with dominating antiferromagnetic exchange reveal strong geometrical frustration, while, in the sulfides and selenides, ferromagnetic exchange becomes important. ZnCr_2S_4 and ZnCr_2Se_4 are strongly frustrated due to competing antiferromagnetic and ferromagnetic interactions, with the ground state still being antiferromagnetic [16]. At low temperatures, the

geometrically frustrated oxide spinels exhibit magnetization plateaus as function of an external magnetic field, with a three-up one-down spin configuration [17–19]. Theoretically, it has been suggested that these plateaus are stabilized by lattice distortions [20]. Indeed, in high-field ultrasonic experiments on CdCr_2O_4 , these magnetization plateaus are also seen as broad strain plateaus [21], indicating a stable structural configuration. In contrast, the high-field magnetization of bond-frustrated ZnCr_2S_4 and ZnCr_2Se_4 is rather smooth [22,23], but nevertheless these compounds show plateaulike anomalies in high-field ultrasound experiments.

In this Letter, we report new effects in the multiferroic spinel CoCr_2O_4 evidenced by anomalies in the ultrasound propagation in magnetic fields up to 62 T. CoCr_2O_4 crystallizes in the normal cubic $Fd\bar{3}m$ structure with Co^{2+} (electronic configuration $3d^7$, spin $S = 3/2$) and Cr^{3+} ($3d^3$, $S = 3/2$) ions occupying tetrahedral and octahedral sites, respectively. A collinear ferrimagnetic spin order sets in below $T_C \approx 95$ K, followed by a transition into an incommensurate (IC) long-range conical-spiral state at $T_s \approx 26$ K [24–26]. Reports on the magnetic structure between T_C and T_s are controversial. Menyuk *et al.* [24] and Tomiyasu *et al.* [25] report on the short-range order of the spiral state. In contrast, Chang *et al.* [26] found a well-defined long-range ordered IC conical spin state. On further lowering the temperature, an incommensurate-to-commensurate (C) lock-in transition occurs at $T_l \approx 14$ K [9,25–27]. The true spin configuration of this C-spiral phase below T_l remains to be clarified and is a matter of controversy [28]. A further remarkable property of CoCr_2O_4 is its multiferroicity. The coupling of charge

and spin leads to anomalies of the dielectric constant at the magnetic transitions [6,29,30]. Importantly, a spontaneous dielectric polarization appears only in the IC-spiral state below T_s [6], indicating a low-symmetry phase. Multiferroicity in CoCr_2O_4 was assigned to the inverse Dzyaloshinskii-Moriya (DM) interaction based on the fact that the observed modulations of the spins and the lattice have the same wave vector [31].

From sound-velocity measurements in solids, detailed information on the elastic constants can be derived. Besides structural phase transitions, ultrasound experiments probe, e.g., spin-phonon coupling, orbital degrees of freedom, or charge order [32], and we observe corresponding anomalies at the transitions into the commensurate and incommensurate-spiral-spin states. Upon increasing the external magnetic field in the commensurate-spiral ground state, a clear transition into a new high-field phase is found. This phase remains metastable upon decreasing the magnetic field down to a lower critical field, below which the incommensurate-spiral ground state is recovered. We propose that this new high-field phase is characterized by a restored (possibly cubic) symmetry as a result of the external magnetic field overcoming the effects of the inverse DM interactions. In this scenario, only the longitudinal (collinear) component of the magnetization remains ordered, while the transverse component becomes disordered.

High-quality CoCr_2O_4 single crystals were grown by chemical transport. The phase purity of the samples was checked by x-ray analysis. The elastic properties were studied by measurements of the velocity and attenuation of longitudinal waves with wave vector \mathbf{k} and polarization \mathbf{u} parallel to the $\langle 001 \rangle$ and $\langle 111 \rangle$ axes, which for a cubic crystal correspond to the elastic constants c_{11} and $c_L = 1/3(c_{11} + 2c_{12} + 4c_{44})$, respectively. The measurements in static magnetic fields up to 14 T were performed for temperatures between 1.5 and 300 K. A phase-sensitive detection technique based on a pulse-echo method [33] was used. Further ultrasound and magnetization measurements were performed in pulsed magnetic fields up to 62 T with a rise time of 35 ms and a pulse duration of 150 ms in the temperature range from 1.5 to 20 K. The magnetic properties were also studied using a SQUID magnetometer (Quantum Design MPMS-5) in static fields up to 5 T.

In Fig. 1, the temperature dependencies of the zero-field-cooled χ_{ZFC} and field-cooled χ_{FC} susceptibilities for CoCr_2O_4 measured in a field of 10 mT applied along the $\langle 001 \rangle$ axis are shown. Both χ_{ZFC} and χ_{FC} exhibit a steplike increase at the ferrimagnetic transition at $T_C \approx 95$ K and pronounced anomalies at the transition into the incommensurate-spiral-spin state at $T_s \approx 27$ K and at the IC-to-C lock-in transition at $T_l \approx 14$ K. Below T_l , a strong magnetization irreversibility and a history dependence appear due to the first-order magnetic phase transition at T_l in agreement with neutron-diffraction data [26]. At temperatures above 250 K, the magnetic susceptibility

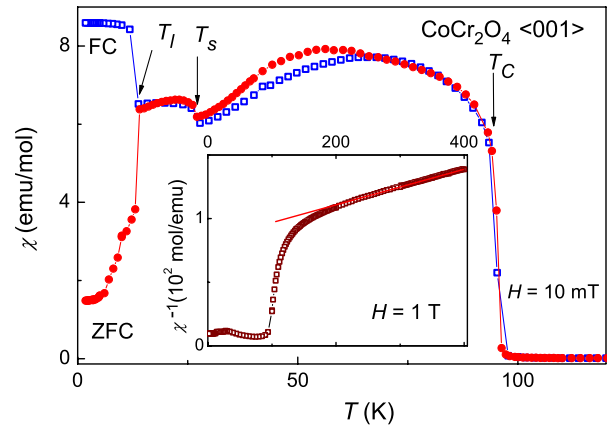


FIG. 1 (color online). Temperature dependence of the ZFC (full circles) and FC susceptibilities (open squares) for CoCr_2O_4 measured in a field of 10 mT applied along the $\langle 001 \rangle$ axis. Inset: Temperature dependence of the inverse susceptibility measured in a field of 1 T. The solid line marks a Curie-Weiss fit at high temperatures.

follows a Curie-Weiss law with an asymptotic Curie-Weiss temperature $\theta_{\text{CW}} \approx -600$ K. Deviations from the Curie-Weiss behavior emerge already below 250 K, indicating the appearance of spin correlations (see the inset of Fig. 1). The high ratio of $|\theta_{\text{CW}}|/T_C \approx 6$ points toward significant magnetic frustration due to competing $BB(\text{Cr}-\text{Cr})$, $AB(\text{Co}-\text{Cr})$, and $AA(\text{Co}-\text{Co})$ exchange interactions that establish the spiral-spin configuration at low temperatures [34,35].

In Fig. 2, the sound velocity v (right scale) and its relative change $\Delta v/v_0$ (left scale), measured in different static magnetic fields for ultrasound waves propagating along the $\langle 111 \rangle$ axis, are presented as a function of

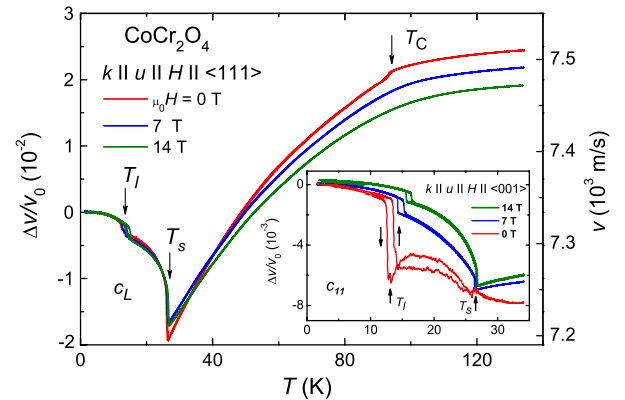


FIG. 2 (color online). Temperature dependencies of the sound velocity v (right scale) and its relative change $\Delta v/v_0$ (left scale) for CoCr_2O_4 measured in different static magnetic fields for ultrasound waves propagating along the $\langle 111 \rangle$ axis. The inset shows $\Delta v/v_0$ for waves propagating along the $\langle 001 \rangle$ axis at low temperatures for cooling and warming cycles. The arrows mark the magnetic phase transitions T_C , T_s , and T_l observed at the zero magnetic field.

temperature. In the high-temperature cubic phase and in the zero magnetic field, $v \sim 7.5$ km/s decreases with increasing fields and, contrary to what is observed in most solids, decreases toward lower temperatures. At $T_C = 95$ K, v exhibits a small anomaly only, indicative for a weak coupling of the longitudinal sound waves to the collinear spin structure which evolves below T_C . Between T_C and the transition into the IC spiral-spin state at T_s , the lattice softening is reflected by a considerable reduction of $\Delta v/v_0$ of about 4%. A similar strong softening was reported for the related frustrated spinel compounds CdCr_2O_4 and $\text{ZnCr}_2\text{S}(\text{Se})_4$ [21–23] when entering the states with incommensurate spin configurations, which is accompanied by a structural transformation from cubic to tetragonal symmetry driven by a spin-Jahn-Teller effect [17,19,36]. At T_s , v shows a deep minimum followed by a steplike increase below T_s . On further decreasing the temperature, v exhibits a third anomaly at the transition into the commensurate state at T_l with a hysteresis on cooling and warming (see the inset in Fig. 2 for the elastic constant c_{11}), confirming the first-order nature of this phase transition. With an increasing magnetic field, the anomaly in the sound velocity at T_l shifts to higher temperatures, whereas the position of the anomaly at T_s remains unchanged (see the inset of Fig. 2).

We now turn to the results obtained in high magnetic fields that are presented in Fig. 3, where the sound velocity v , the sound attenuation $\Delta\alpha$, and the magnetization M are shown as functions of an applied pulsed magnetic field for different temperatures. At the lowest temperature of 1.5 K, the sound velocity increases monotonically with the magnetic field in a fully reversible manner. This reflects the field dependence of the sound velocity in the spiral-spin configuration, which is stable up to 62 T. At this temperature, the sound attenuation changes nonmonotonically with the field, decreasing up to 25 T, passing through a broad minimum, and increasing again toward higher fields without hysteresis. In contrast, at 6.25 K, a steplike increase in the sound velocity and a kinklike sharp maximum in the attenuation are found in a field $H_t = 55$ T, signaling the transition into a new high-field phase with significantly enhanced stiffness. We want to point out that the absolute value of the sound velocity in this state is close to $v(T_C)$, the velocity in the vicinity of T_C (see Fig. 2). This hints toward a similarity of the new high-field phase and the high-symmetry phase at higher temperatures. Astonishingly, upon reducing the applied field, the ground state is not recovered at the same magnetic-field values H_t , but the high-field phase remains metastable down to a lower transition field and seems to persist even to zero fields at 6.25 K. At 6.25 K, the field dependence of the sound attenuation on increasing fields up to H_t resembles the behavior at 1.5 K. For fields above H_t , the slope of the attenuation changes sign, and, for the down sweep, a maximum appears close to 25 T. On further increasing

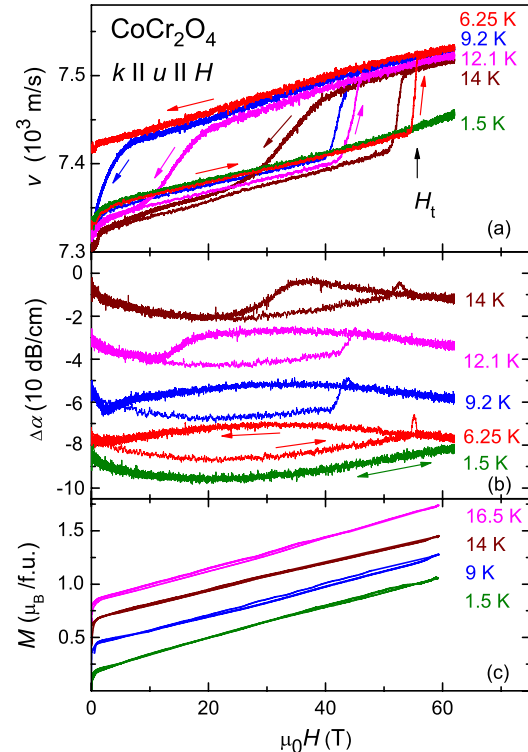


FIG. 3 (color online). (a) Sound velocity v , (b) sound attenuation $\Delta\alpha$, and (c) magnetization M in CoCr_2O_4 at different temperatures vs the magnetic field applied along the $\langle 111 \rangle$ direction. The data for the up and down field sweeps are shown. The vertical arrow marks the transition field H_t at 6.25 K. The tilted arrows indicate the field sweep directions. The curves in (b) and (c), except that for 1.5 K, are shifted along the vertical axis for clarity.

temperatures, the transition field H_t reaches a minimum close to 42 T at 9 K and increases again to 61 T at 16.5 K. The transitions at temperatures above 9.2 K are characterized by a large hysteresis which becomes narrower on increasing temperature.

Using our ultrasound results, we constructed the low-temperature phase diagram of CoCr_2O_4 , which is plotted in Fig. 4. The transition from the collinear ferrimagnetic (FiM) phase to the IC phase at $T_s = 27$ K is almost field independent. The subsequent transition to the C spiral phase increases linearly with the external magnetic field at least up to 14 T. For fields higher than 42 T, a high-field phase appears. This high-field phase is metastable and prevails with the decreasing field down to 0 T, close to 6 K. At the lowest temperatures and at least up to 62 T, the commensurate-spiral phase is the stable ground state.

The magnetization M measured at several temperatures below T_s shows an initial sharp ferromagneticlike increase in fields below 2 T followed by a strictly linear dependence at higher fields. The ferromagnetic component has a value of $0.17 \mu_B/\text{f.u.}$ and changes slightly with increasing temperature from 1.5 to 27 K. Note that, between 1.5 and 16.5 K, M increases with an almost constant slope at

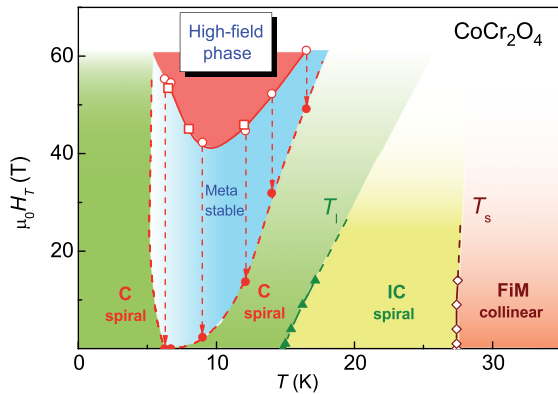


FIG. 4 (color online). Schematic low-temperature phase diagram of CoCr_2O_4 . The phase boundaries between the metastable phase and the helical C phase detected in the pulsed experiments are shown by dashed lines. The arrows connect the boundaries observed in increasing fields (open symbols) and decreasing fields (solid circles). The open squares and circles refer to the ultrasound data measured in the pulsed fields applied along the $\langle 001 \rangle$ and $\langle 111 \rangle$ directions, respectively.

high fields. Hence, the magnetization evidences no sign of magnetic phase transitions, neither in passing the commensurate-to-incommensurate phase boundary nor in entering the high-field phase regime detected in the ultrasound experiments [see Fig. 2 and Figs. 3(a) and 3(b)]. Similarly smooth magnetization curves have been reported for the multiferroic system $\text{Ni}_3\text{V}_2\text{O}_8$, when the external magnetic field is perpendicular to the magnetic easy axis [37,38], while distinct magnetization steps are revealed parallel to the easy axis [39]. In CoCr_2O_4 , where the magnetic anisotropy is by far smaller than in $\text{Ni}_3\text{V}_2\text{O}_8$, we do not even expect any magnetization steps for other orientations of the external field because the propagation of the spiral always follows the external field. Only nuclear magnetic resonance or neutron scattering at high fields could provide essential information about changes of the transverse magnetization component suggested in the proposed scenario.

This absence of concurrent features in the magnetization at first glance suggests that the high-field anomaly in the sound velocity is connected to purely structural changes. On the other hand, the steplike increase of the sound velocity to a value characteristic for the high-symmetry paramagnetic or collinear phases suggests an abrupt magnetic first-order transition from the low-symmetry spin-spiral state to a high-symmetry possibly collinear spin state. In a rather narrow temperature regime, this high-field phase remains metastable even in the zero external field.

A similar field-induced effect with an increase of the lattice stiffness was demonstrated recently for the frustrated $\text{ZnCr}_2\text{S}(\text{Se})_4$ spinels and ascribed to the recovery of a high lattice symmetry [22,23]. However, in these systems, a clear structural symmetry reduction is present in the zero field. Also, in the frustrated oxide spinels, e.g.,

HgCr_2O_4 and CdCr_2O_4 , the release of the geometrical frustration by the magnetic field is accompanied by a structural transformation from tetragonal to cubic [17,19].

Given the occurrence of a spontaneous polarization in CoCr_2O_4 , we suppose that, at low temperatures and zero magnetic field, the lattice symmetry of CoCr_2O_4 must already be lower than cubic.

The external high magnetic field will increase the Zeeman energy of the Cr^{3+} ($g = 2$) and Co^{2+} ($g = 2.23$) [40,41] ions to about 7.3 and 8.1 meV, respectively, when using the minimal magnetic transition field of $H_T = 42$ T at 9 K. Theoretical estimates for the energy of the nearest-neighbor exchange interactions obtained from LSDA + U calculations are $J_{\text{Cr-Co}} = 3.6\text{--}4.3$ meV, $J_{\text{Cr-Cr}} = 1.3\text{--}3.3$ meV, and $J_{\text{Co-Co}} = 0.5\text{--}0.23$ meV (including the spin values) [35]. Taking into account the number of interacting nearest neighbors, only the Cr-Co interaction is significantly larger than the magnetic-field energy. On the other hand, the Co-Co interaction is easily overcome by the external field, while Cr-Cr interaction is comparable to the observed transition field. Thus, the external magnetic field changes the balance between the competing exchange interactions and finally overrules the interaction responsible for the reduced symmetry.

In addition, the leading contribution of spin-lattice coupling and the origin of multiferroicity was assigned to arise from the inverse DM interaction and not from the conventional symmetric exchange striction [31]. The puzzling fact that we do not observe any anomaly in the field dependence of the magnetization $M(H)$, which is a measure of the longitudinal component of the magnetization, may become understandable by assuming that only the transverse component is affected in the high-field transition. With regard to the long debate on the short-range and long-range magnetic order of the spiral component of the magnetization [25,26], we suggest that sufficiently strong external magnetic fields destroy the spiral order of the transverse magnetization component. This results in a quasiparamagnetic state, while the longitudinal component resides in collinear long-range order. Concomitantly to the breakdown of the transverse spiral order, the effects of the inverse DM interaction are canceled and the structure is released into presumably purely cubic symmetry. The high-field state remains metastable on lowering the field until the transverse magnetization is large enough to reestablish the spiral state. The neutron-scattering results [24–26] indicate that short-range and long-range ordered spirals are of similar ground-state energy, providing the precondition for the metastability.

In conclusion, the present high-field studies of CoCr_2O_4 in magnetic fields up to 62 T reveal a remarkable phase diagram and unconventional magnetoelastic effects: While the transition from the paramagnetic into the collinear ferrimagnetic state reveals weak anomalies only, the lattice softens considerably on approaching the low-temperature

spiral-spin phase. The softening is continuous as a function of temperature but is abrupt as a function of the magnetic field. The main result of our study is a fascinating island of stability of a field-induced phase at low temperatures with a minimal critical field of 42 T at 9.5 K. We speculate that this is a high-symmetry—probably cubic—phase, where the long-range order of the transverse component of the magnetization is suppressed concomitantly with the exchange-driven structural distortions.

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