Charge order breaks time-reversal symmetry in CsV₃Sb₅

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The recently discovered vanadium-based kagome metals ⁴AV₃Sb₅ (A = K, Rb, Cs) exhibit superconductivity at low-temperatures and charge density wave (CDW) order at high-temperatures. A prominent feature of the charge ordered state in this family is that it breaks time-reversal symmetry (TRSB), which is connected to the underlying topological nature of the band structure. In this work, a powerful combination of zero-field and high-field muon-spin rotation/relaxation is used to study the signatures of TRSB of the charge order in CsV₃Sb₅, as well as its anisotropic character. By tracking the temperature evolution of the in-plane and out-of-plane components of the muon-spin polarization, an enhancement of the internal field width sensed by the muon-spin ensemble was observed below TRSB = TCDW ≃ 95 K. Additional increase of the internal field width, accompanied by a change of the local field direction at the muon site from the ab-plane to the c-axis, was detected below T* ≃ 30 K. Remarkably, this two-step feature becomes well pronounced when a magnetic field of 8 T is applied along the crystallographic c-axis, thus indicating a field-induced enhancement of the electronic response at the CDW transition. These results point to a TRSB in CsV₃Sb₅ by charge order with an onset of ≃ 95 K, followed by an enhanced electronic response below ≃ 30 K. The observed two-step transition is discussed within the framework of different charge-order instabilities, which, in accordance with density functional theory calculations, are nearly degenerate in energy.

I. INTRODUCTION

The emergence of metallic kagome materials featuring an intricate structural lattice and rich diversity of quantum phases has reinvigorated the quest for finding materials with topological phases built from strongly interacting electrons.¹–³ This led to the discovery of a vanadium-based kagome metal family ⁴AV₃Sb₅ (A = K, Rb, Cs),⁴–⁶ which was reported to feature a metallic, topological phase at high temperature, anomalous transverse transport properties including the anomalous Hall effect,⁷ anomalous Nernst effect,⁸ unconventional planar Hall effect,⁹ a transition to a highly tunable unconventional superconducting state at low temperatures,¹⁰–¹⁵ and time-reversal symmetry breaking (TRSB) charge order. The TRSB isotropic 2×2 charge density wave (CDW) order was first suggested by magnetic-field-based scanning tunneling microscopy data¹⁶–²¹ and was later widely discussed theoretically.²²–²⁵,²⁸ Several groups observed a reduced CDW symmetry obtained by a 4×1 charge modulation in the CDW state,²⁶ which would reduce the rotational symmetry from sixfold C₆ to twofold C₂.

The combination of zero-field (ZF) and high transverse-field (TF) muon-spin rotation/relaxation (μSR) experiments has provided direct evidence for TRSB below the onset of charge order in KV₃Sb₅.¹⁴ Similarly, the appearance of spontaneous fields below the charge ordering temperature was also reported for RbV₃Sb₅.²⁷ ZF-μSR experiments on the sister compound CsV₃Sb₅ have reported the onset of the TRSB state at T_{TRSB} ≃ 70 K,²⁸ which is lower than the CDW transition temperature T_{CDW} > 95 K. A more recent ZF-μSR study of CsV₃Sb₅ reported the appearance of spontaneous fields below 50 K.²⁹ In contrast to ZF-μSR experiments, Kerr effect measurements reveal the emergence of a TRSB signal in CsV₃Sb₅ exactly at T_{CDW}.³⁰ Consequently, the determination of the true onset of spontaneous fields in CsV₃Sb₅ as well as their in-plane and the out-of-plane anisotropy are of paramount importance as they should intimately relate to the mechanism of charge order.

In this paper, we utilize the combination of ZF and TF-μSR techniques to probe the μSR relaxation rates in CsV₃Sb₅ as a function of temperature, field, and angle α between the in-plane component of the muon-spin polarisation and the crystallographic a–axis. The main observation is a two-step increase of the internal field width sensed by the muon-spin ensemble. It consists of a noticeable enhancement at T_{TRSB} ≃ 95 K, corresponding to the CDW ordering temperature T_{CDW}, followed by a stronger increase below T* ≃ 30 K. An applied magnetic field of 8 T along the crystallographic c–axis further enhances the magnetic response below T_{CDW}, leading to a much more pronounced two-step increase of the internal field width. Furthermore, the local field at the muon site lies within the a–a–plane of the crystal in the temperature range from T_{CDW} to T*. Below T*, the internal field also acquires an out-of-plane component. The absence of the in-plane anisotropy of the internal fields down to ≃ 3 K was also detected, while the out-of-plane anisotropy remains strong. Our results provide evidence...
for time-reversal symmetry-breaking in CsV₃Sb₅ at the onset of charge order, as well as a non-trivial temperature evolution of the electronic response within the charge ordered state. More generally, these results indicate a strong interplay between magnetic and charge channels in this kagome material.

The paper is organized as follows: Section II describes the sample preparation procedure and details of the μSR experiment. Details of the zero-field and transverse-field μSR data analysis process are given in Section II.E. The results of the zero-field and 8 T transversal-field μSR experiments are discussed in Section III. Conclusions follow in Section IV.

II. EXPERIMENTAL DETAILS

A. Sample preparation and characterization

Single crystals of CsV₃Sb₅ were grown following the procedure described in Ref. 31. Single crystals with dimensions of ≃ 3 × 3 × 1 mm³ were used. As demonstrated in Ref. 31, the CsV₃Sb₅ single crystals possess an obviously hexagonal shape, which allows one to easily distinguish the main crystallographic axes (a and c).

The X-ray Laue diffraction images of the studied crystals demonstrate the single crystallinity of the material and confirm the correspondence of the main crystal axes to the sample shape. The Laue images for six individual crystals used in "in-plane" rotation experiments are shown in the Supplementary Information. Based on the results of the Laue studies, the crystals were aligned along a and c axes and mounted on a specially constructed μSR sample holder (see Sec. II C and the Supplementary Information, for further details).

The superconducting transition temperature Tc was determined by means of ac susceptibility and was found commonly for all crystals to be Tc ≃ 2.7 K, in agreement with previously published data. The ac susceptibility curves for two sets of crystals used in zero-field and transverse-field μSR experiments are presented in the Supplementary Information.

B. Muon spin rotation/relaxation experiments

The muon spin rotation/relaxation (μSR) experiments were carried out at the πM3 and πE3 beam lines by using the GPS, Ref. 34, and HAL-9500 spectrometers (Paul Scherrer Institute, Switzerland). The μSR measurements were performed at temperatures ranging from ≃ 3 to 300 K. The 100% spin-polarized "surface" muons with a momentum of pµ ≃ 28.6 MeV/c were implanted into the CsV₃Sb₅ crystals along the c-axis [see Fig. 1 (a)]. Muons thermalize rapidly without a significant loss of their initial spin polarization and stop in matter at the depth of about 0.15 g/cm². For CsV₃Sb₅, with the density of about 5 g/cm³, this corresponds to a depth of ≃ 0.3 mm. With the CsV₃Sb₅ crystal thickness of ≃ 1 mm, all "surface" muons stop in the sample, so the use of degraders, as it is required in μSR studies of thin single-crystal samples, was not necessary.

C. ZF-μSR experiments

Experiments with zero applied field were performed at the GPS spectrometer. Measurements were made by varying two angles: β, the angle between the initial muon-spin polarization Pµ and the muon momentum pµ;
and \( \alpha \), the angle between the crystal’s \( a \)-axis and the in-plane component of the muon-spin polarization \( \mathbf{P}_{\mu}^{ab} \) [see Fig. 1 (a)]. Both "in-plane" and "out-of-plane" ZF-\( \mu \)SR experiments were conducted. In the "in-plane" rotation experiments, the angle \( \beta \) was kept at the maximum value allowed by the spin-rotator setup at the \( \pi \)M3 beam-line (\( \beta \approx 60^\circ \), Ref. 34). Temperature scans were performed for three different values of \( \alpha = 0^\circ, 30^\circ \) and \( 60^\circ \). In these experiments, the time evolution of \( \mathbf{P}_{\mu}^{ab} \) component of the muon-spin polarization [Fig. 1 (a)] was accessed. The "out-of-plane" experiments were conducted with \( \alpha \) and \( \beta \) set to \( 30^\circ \) and \( 5^\circ \), respectively. The temperature evolution of \( \mathbf{P}_{\mu}^{ac} \) component of the muon-spin polarization was measured. Note that \( \beta = 5^\circ \) corresponds to the smallest possible muon-spin rotated angle at the GPS instrument.34

In order to vary the angle \( \alpha \), a special sample holder was constructed [Fig. 1 (b)]. It consists of an aluminum support plate and two sample mounting rings. Each sample ring has 36 holes, which allows for rotation with a \( 10^\circ \) step relative to the support plate. The \( a- \) and \( c- \)-axis-aligned Cs\( \text{V}_3\text{Sb}_5 \) crystals were glued on a 25 \( \mu \)m aluminum foil, which was further attached to one of the aluminum sample rings. The second ring was covered by a 25 \( \mu \)m thin layer of Kapton in order to prevent the crystals from falling down inside the cryostat. Note that the Kapton and aluminum foils are fully transparent for the "surface" muons, which allows us to use the advantage of the so-called "Veto" mode. The "Veto" mode rejects the muons missing the sample and, as a consequence, reduces the background of the \( \mu \)SR response to nearly zero (see Ref. 34, for a detailed explanation of the "Veto" mode principle). Photos of six Cs\( \text{V}_3\text{Sb}_5 \) crystals mounted on the muon sample holder and the background estimate are given in the Supplementary Information.32

D. TF-\( \mu \)SR experiments

Experiments with the magnetic field applied transversal to the initial muon-spin polarisation (TF-\( \mu \)SR experiments) were performed at the HAL-9500 spectrometer. The initial muon-spin polarization was set perpendicular to the muon-momentum \( [\beta \approx 90^\circ \), Fig. 1 (a)]. The external magnetic field, \( B_{\text{ext}} = 8 \) T, was applied parallel to the muon-momentum \( (B_{\text{ext}} \parallel P_\mu) \) i.e. parallel to the crystallographic \( c- \)-axis \( (B_{\text{ext}} \parallel c) \). In these experiments the time evolution of the \( \mathbf{P}_{\mu}^{ab} \) component of the

![Figure 2](https://example.com/figure2.png)

**FIG. 2:** (a)–(b) Temperature dependencies of the Gaussian Kubo-Toyabe \( [\sigma_{\text{GKT}}, \text{panel (a)}] \) and exponential \( [\lambda, \text{panel (b)}] \) relaxation rates obtained from fits of the \( \mathbf{P}_{\mu}^{ab}(t) \) components of the muon-spin polarization in ZF-\( \mu \)SR experiments. The angles \( \alpha \) and \( \beta \) were set to \( 30^\circ \) and \( 60^\circ \), respectively. (c)–(d) The same as in panels (a) and (b), but for \( \mathbf{P}_{\mu}^{ac}(t) \) and \( \beta = 5^\circ \). The dashed lines are linear fits in the temperature regions: \( 5 \text{ K} \leq T \leq 30 \text{ K}, 30 \text{ K} \leq T \leq 95 \text{ K}, \) and \( 95 \text{ K} \leq T \leq 300 \text{ K} \). The broad gray and violet lines in panels (a) and (c) represent the CDW ordering temperature \( T_{\text{CDW}} \) and the characteristic temperature \( T^\star \). The broad lines in panels (b) and (d) represent the TRSB transition temperature \( T_{\text{TRSB}} \) and \( T^\star \), respectively.
muon-spin polarization [Fig. 1 (a)] was accessed. Crystals were mounted on the sample-holder made of 99.999% pure silver, and can be seen in a photo shown in the Supplementary Information.32

E. μSR data analysis procedure

The zero-field μSR spectra were fitted using the Gaussian Kubo-Toyabe (GKT) relaxation function,35,36 describing the nuclear moment response, multiplied by an additional exponential term:

\[ A_i^{ZF}(t) = A_{0,i}^{ZF} \left[ \frac{1}{3} + \frac{2}{3}(1 - \sigma_{GKT}^2 t^2) e^{-\sigma_{GKT}^2 t^2/2} \right] e^{-\lambda t}. \]  

Here, \( A_{0,i}^{ZF} \) is the initial asymmetry of the \( i \)-th positron detector at \( t = 0 \), \( \sigma_{GKT} \) is the GKT relaxation rate, and \( \lambda \) is the exponential relaxation rate. Note that Eq. 1 is widely used to analyze the ZF-μSR data in most TRSB μSR studies.14,37–40

The TF-μSR data were analyzed as:

\[ A_i^{TF}(t) = A_{0,i}^{TF} \cos(\gamma_i B_{\text{int}} t + \phi_i) e^{-\sigma_i^2 t^2/2} e^{-\lambda t}. \]  

Here \( B_{\text{int}} \) is the internal field at the muon site, \( \phi_i \) is the initial phase of the muon-spin ensemble, \( \gamma_i = 2\pi \times 135.5 \text{ MHz/T} \) is the muon gyromagnetic ratio, and \( \sigma_i \) is the Gaussian relaxation rate.

In the above Eqs. 1 and 2, \( \sigma_{GKT} \) and \( \sigma \) relaxation rates mainly account for the nuclear moment contribution, which is assumed to be static within the μSR time window. As discussed previously for KV\(_3\)Sb\(_5\) and RbV\(_3\)Sb\(_5\), the exponential relaxation rate \( \lambda \) is mostly sensitive to the temperature dependence of the electronic contribution to the muon spin relaxation.14,27 One cannot exclude, however, subtle effects owing to changes in the electric field gradients in the charge ordered state.51 The details of the ZF- and TF-μSR data analysis procedure are discussed in the Supplementary Information.32

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

The parameters obtained from the analysis of ZF- and TF-μSR data are summarized in Figs. 2, 3, and 4.

Figure 2 compares the results of the "in-plane" and "out-of-plane" ZF-μSR experiments conducted at \( \alpha = 30^\circ \). The left and right columns of Fig. 2 represent the parameters obtained from the fits of the \( P_{\mu}^{ab}(t) \) and \( P_{\mu}^{c}(t) \) components of the muon-spin polarization, respectively.

Figure 3 shows the temperature evolution of the fit parameters obtained from the 8 T TF-μSR experiments. Panels (a), (b), and (c) refer to the temperature dependencies of \( \sigma \), \( \lambda \), and the experimental muon Knight shift \( (B_{\text{int}} - B_{\text{ext}})/B_{\text{ext}} \), respectively.

Figure 4 shows the temperature dependencies of the Gaussian Kubo-Toyabe [panel (a)] and the exponential [panel (b)] relaxation rates obtained in the "in-plane" rotation ZF-μSR set of experiments. The time evolution of the in-plane component of the muon-spin polarization \( P_{\mu}^{ab}(t) \) was analyzed. The closed circles, open circles and closed triangles depict the data taken at \( \alpha = 0^\circ \), 30°, and 60°, respectively.

The experimental results presented in Figs. 2, 3 and 4 are closely related to each other and display distinct features, which will be discussed in the following Sections.
A. Two-step feature and out-of-plane anisotropy of the internal field

Figure 2 shows that three of four ZF relaxation rates, namely $\lambda$ in the $P^c$ set of experiments ($\lambda^c$) and the two Kubo-Toyabe relaxation rates from the $P^{ab}$ and $P^c$ sets of data ($\sigma_{\text{GKT}}^{ab}$ and $\sigma_{\text{GKT}}^c$) display a sudden change across $T_{\text{CDW}} \simeq 95$ K (indicated by the thick grey lines in all four panels of Fig. 2). Namely, both $\sigma_{\text{GKT}}$ suddenly increase, while $\lambda$ decreases when crossing the CDW transition temperature. The absolute value of the jump, as estimated from the linear fits of the relaxation rate data above and below $T_{\text{CDW}}$, was found to be the same (within the experimental uncertainty) and corresponds to $\simeq 0.0075(20) \text{ ms}^{-1}$ [see Figs. 2 (a), (c), and (d)]. The appearance of a step-like change of both $\sigma_{\text{GKT}}$ and $\lambda$ agrees with the first-order nature of the CDW transition of CsV$_3$Sb$_5$, reported in Refs. 4–6,16,19–21,42–45

Upon lowering the temperature below $T_{\text{CDW}}$, two different slopes above and below the characteristic temperature $T^* \simeq 30$ K are detected for $\lambda^c(T)$ [Fig. 2 (d)]. At the same time, $\lambda^{ab}(T)$ remains at zero for $T > T^*$ and increases with decreasing temperature below $T^*$ [Fig. 2 (b)]. In a simplified description, the finite experimental relaxation rate observed in the $P^c$ experiments accounts for the width of the field distribution in the $ab$-plane ($\Delta B^\perp$), while a finite relaxation along $P^c$ is associated with the field components from the $ac$- and/or $bc$-planes ($\Delta B^\parallel$). Accounting for the experimental data presented in Figs. 2 (b), 2 (d), and 4 (b), this implies that for $T^* \lesssim T < T_{\text{CDW}}$ the internal field on the muon position has only $B^{\parallel}$ components, while for $T \lesssim T^*$ both $B^{\parallel}$ and $B^\perp$ components are present.

The increase of the exponential contribution to the internal field width is also accompanied by a non-monotonic temperature dependence of the Gaussian contribution to the internal field width. Below $T_{\text{CDW}} \simeq 95$ K, the Gaussian relaxation rate changes in two steps. In the region $T^* \lesssim T \lesssim T_{\text{CDW}}$, $\sigma^{ab}_{\text{GKT}}(T)$ decreases, while $\sigma^c_{\text{GKT}}(T)$ stays nearly constant with decreasing temperature. Below $T^* \simeq 30$ K, the temperature dependence of both relaxation components change slope and begin to increase.

While the increase of the exponential relaxation $\lambda^c(T)$ of CsV$_3$Sb$_5$ is consistent with the onset of time-reversal symmetry-breaking at $T_{\text{CDW}}$ ($T_{\text{TRSB}} \simeq T_{\text{CDW}}$), high-field $\mu$SR experiments are essential to confirm this effect, as the ZF and weak TF-$\mu$SR data can be subtly affected by the onset of different charge orders even without the presence of TRSB. Following Ref. 14, the application of a high magnetic field leads to a strong enhancement of the electronic contribution to the relaxation rate. Comparison of the relaxation rates obtained in ZF- and TF-$\mu$SR experiments confirms that this is actually the case. Indeed, $\sigma^{ab}(T)$ measured at $B_{\text{ext}} = 8$ T [Fig. 3 (a)] reproduces the temperature evolution of $\sigma_{\text{GKT}}$ collected in ZF-$\mu$SR studies [Fig. 2 (a)]. The absolute values of both $\sigma^{ab}$ and $\sigma_{\text{GKT}}^{ab}$ stay almost the same within 10-15%. In contrast, the exponential component is strongly affected by the applied field. There is a factor of $\sim 1.5 - 2$ enhancement of $\lambda^{ab}(T)$ in the $T \lesssim 30$ K region and a full recovery of the $\lambda^{ab}$ component for temperatures between 30 K $\lesssim T \lesssim 95$ K. This gives rise to a well-pronounced two-step feature at high fields. Note that such a two-step increase of the relaxation rate is also observed in the sister compound RbV$_3$Sb$_5$.27

To estimate the onset temperatures associated with this two-step transition, the TF $\lambda^{ab}(T)$ was fitted with a double-stage power-law functional form. While this is not a microscopically derived function, it allows us to gain quantitative insight about the temperatures involved. The fit function assumes that each state is characterised by its own transition temperature ($T_{\text{TRSB}}$ and $T^*$), the enhancement of $\lambda(T)$ due to spontaneous magnetic fields ($\Delta \lambda_1$ and $\Delta \lambda_2$), and the power-law exponent ($n_1$ and $n_2$):

$$\lambda(T) = \begin{cases} 0, & T > T_{\text{TRSB}} \\ \Delta \lambda_1 \left[ 1 - \left( \frac{T}{T_{\text{TRSB}}} \right)^{n_1} \right], & T^* < T < T_{\text{TRSB}}, \\ \Delta \lambda_1 \left[ 1 - \left( \frac{T}{T^*} \right)^{n_1} \right] + \Delta \lambda_2 \left[ 1 - \left( \frac{T}{T^*} \right)^{n_2} \right], & T < T^*. \end{cases}$$

(3)

The solid line in Fig. 3 (b) represents the results of the fit. The parameters are: $T_{\text{TRSB}} = 94.9(1.4)$ K, $\Delta \lambda_1 = 0.0022(2) \text{ ms}^{-1}$, $n_1 = 5.3(1.5)$ and $T^* = 31.4(2.8)$ K, $\Delta \lambda_2 = 0.0014(3) \text{ ms}^{-1}$, $n_2 = 3.7(2.5)$ for the first and the second step, respectively.

The combination of ZF-$\mu$SR and high-field TF-$\mu$SR results on CsV$_3$Sb$_5$ provides an indication of time-reversal symmetry-breaking below the onset of charge order $T_{\text{TRSB}} = T_{\text{CDW}} \simeq 95$ K. This agrees well with the previous reports on KV$_3$Sb$_5$14 and RbV$_3$Sb$_5$,27 indicating that the TRSB effect is strongly connected to the charge-density wave transition for all three members of this kagome metal family. It has to be mentioned, however, that the effects of TRSB in CsV$_3$Sb$_5$ are much less pronounced than for the sister compounds KV$_3$Sb$_5$14 and RbV$_3$Sb$_5$27 and might easily be overlooked by less precise
TABLE I: The results of the density functional theory (DFT) calculations for CsV$_3$Sb$_5$ obtained in Ref. 46. The columns "Space Symmetry", "Lattice Symmetry", "Rotation Symmetry", and $E_{\text{CDW}} - E_{\text{parent}}$ per f.u. denote the space group of the CDW unit cell, the rotation symmetry along the $c$–axis, and the internal energy difference between the CDW ordered and the parent (non-ordered) states per formula unit, respectively.

| CDW order                          | Space group  | Rotation symmetry | $E_{\text{CDW}} - E_{\text{parent}}$ per f.u. |
|------------------------------------|--------------|-------------------|---------------------------------------------|
| Planar tri-hexagonal               | $P6/mmm$ (#191) | $C_6$ (6-fold)    | $-21$ meV                                   |
| Superimposed tri-hexagonal Star-of-David | $P6/mmm$ (#191) | $C_6$ (6-fold)    | $-17$ meV                                   |
| Staggered tri-hexagonal            | $Fmmm$ (#69)  | $C_2$ (2-fold)    | $-24$ meV                                   |
At the present stage it is difficult to establish a direct relation between the in-plane isotropic internal fields and the symmetry of the CDW. Further theoretical and experimental studies, including the exact determination of the muon-stopping site(s), are required.

IV. CONCLUSION

In conclusion, a combination of zero-field and high transverse-field muon-spin rotation/relaxation experiments were performed on the CsV$_3$Sb$_5$ representative of the kagome superconducting family AV$_3$Sb$_5$ (A =K, Cs, Rb). The in-plane and out-of-plane electronic responses as a function of temperature and magnetic field in the normal state were studied. An enhancement of the width of the internal magnetic field distribution sensed by the muon-spin ensemble was found to coincide with the onset of the charge ordering transition, thus suggesting that the CDW order breaks time-reversal symmetry. A magnetic field of 8 T applied along the crystallographic c-axis further promotes the electronic response below $T_{\text{CDW}}$, leading to a more clearly pronounced two-step increase of the internal field width at the characteristic onset temperatures $T_{\text{FRSB}} = T_{\text{CDW}} \simeq 95$ K and $T^* \simeq 30$ K, respectively. The local fields at the muon stopping site, which are potentially created by loop currents, were found to be confined within the crystallographic $ab$-plane for temperatures between $T_{\text{CDW}}$ and $T^*$, while they possess a pronounced out-of-plane component below $T^*$. Rotation of the crystals around the $c$-axis suggests that the internal field remains isotropic within the kagome plane, in sharp contrast to the highly anisotropic out-of-plane behaviour. Our results indicate a rich electronic response promoted by complex charge order realized in the kagome superconductor CsV$_3$Sb$_5$ and provide useful insights into the nature of the time-reversal symmetry-breaking charge density wave order.

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1 I. Syözı, Statistics of Kagome Lattice. Prog. Theor. Phys. 6, 306 (1951). doi: 10.1143/ptp/6.3.306
2 J.-X. Yin, W. Ma, T. A. Cochran, X. Xu, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian, Z. Song, G. Chang, I. Belopolski, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, S. S. Zhang.
Tsirkin, J.-X. Yin, I. Belopolski, H. Zhou, G. Simutis, S.-S. Zhang, T.A. Cochran, G. Chang, E. Ponomareva, L. Keller, Z. Skrzeczowska, Q. Wang, H. C. Lei, R. Khasanov, A. Amato, S. Jia, T. Neupert, H. Luetkens, and M. Z. Hasan. Tunable anomalous Hall conductivity through volume-wise magnetic competition in a topological kagome magnet. Nat. Commun. 11, 559 (2020).

doi: 10.1038/s41467-020-14325-w

B. R. Ortiz, L. C. Gomes, J. R. Morey, M. Winiarski, M. Bordelon, J. S. Mangum, I. W. H. Oswald, J. A. Rodriguez-Rivera, J. R. Neilson, S. D. Wilson, E. Ertekin, T. M. McQueen, and E. S. Toberer. New kagome prototype materials: discovery of $\text{K}_2\text{V}_3\text{Sb}_5$, $\text{Rb}_2\text{V}_3\text{Sb}_5$, and $\text{Cs}_2\text{V}_3\text{Sb}_5$. Phys. Rev. Materials 3, 094407 (2019).

doi: 10.1103/PhysRevMaterials.3.094407

B. R. Ortiz, S. M. L. Teicher, Y. Hu, J. L. Zuo, P. M. Sarte, E. C. Schueller, A. M. Milinda Abeykoon, M. J. Krogstad, B. R. Ortiz, L. C. Gomes, J. R. Morey, M. Winiarski, M. Bordelon, J. S. Mangum, I. W. H. Oswald, J. A. Rodriguez-Rivera, J. R. Neilson, S. D. Wilson, E. Ertekin, T. M. McQueen, and E. S. Toberer. New kagome prototype materials: discovery of $\text{K}_2\text{V}_3\text{Sb}_5$, $\text{Rb}_2\text{V}_3\text{Sb}_5$, and $\text{Cs}_2\text{V}_3\text{Sb}_5$. Phys. Rev. Materials 3, 094407 (2019).

doi: 10.1103/PhysRevMaterials.3.094407

C. C. Zhao, L. S. Wang, W. Xia, Q. W. Yin, J. M. Ni, Y. Zhuyi Zhang, Zheng Chen, Ying Zhou, Yifang Yuan, H. Yang, Y. Zhang, Z. Huang, Z. Zhao, J. Shi, G. Qian, B. R. Ortiz, S. M. L. Teicher, Y. Hu, J. L. Zuo, P. M. Sarte, E. C. Schueller, A. M. Milinda Abeykoon, M. J. Krogstad, B. R. Ortiz, L. C. Gomes, J. R. Morey, M. Winiarski, M. Bordelon, J. S. Mangum, I. W. H. Oswald, J. A. Rodriguez-Rivera, J. R. Neilson, S. D. Wilson, E. Ertekin, T. M. McQueen, and E. S. Toberer. New kagome prototype materials: discovery of $\text{K}_2\text{V}_3\text{Sb}_5$, $\text{Rb}_2\text{V}_3\text{Sb}_5$, and $\text{Cs}_2\text{V}_3\text{Sb}_5$. Phys. Rev. Materials 3, 094407 (2019).

doi: 10.1103/PhysRevMaterials.3.094407

Y. G. Pan, W. Xia, L. Zhang, K. Yang, X. Mi, A. Wang, Y. Chai, Y. Guo, X. Zhou, and M. He. Magnetoe-Seebeck effect and ambipolar Nernst effect in $\text{Cs}_3\text{V}_5\text{Sb}_5$ superconductor. Phys. Rev. B 104, 180508 (2021).

doi: 10.1103/PhysRevB.104.L180508

L. Li, E. Yi, B. Wang, G. Yu, B. Shen, Z. Yan, and M. Wang. Unconventional Planar Hall Effect in Kagome Metal $\text{K}_2\text{V}_3\text{Sb}_5$. arXiv:2110.07538 (2021).

H. Yang, Y. Zhang, Z. Huang, Z. Zhao, J. Shi, G. Qian, B. Hu, Z. Lu, H. Zhang, C. Shen, X. Lin, Z. Wang, S. J. Pennycook, H. Chen, X. Dong, W. Zhou, and H.-J. Gao. Doping and two distinct phases in strong-coupling kagome superconductors. arXiv:2110.11228 (2021).

C. C. Zhao, L. S. Wang, W. Xia, Q. W. Yin, J. M. Ni, Y. Y. Huang, C. P. Tu, Z. C. Tao, Z. J. Tu, C. S. Gong, H. C. Lei, Y. F. Guo, X. F. Yang, and S. Y. Li. Nodal superconductivity and superconducting domes in the topological Kagome metal $\text{Cs}_3\text{V}_5\text{Sb}_5$. arXiv:2102.08356 (2021).

Zhuyi Zhang, Zheng Chen, Ying Zhou, Yifang Yuan, Shuyang Wang, Jing Wang, Haiyang Yang, Chao An, Lili Zhang, Xianglei Zhou, Yonghui Zhou, Xinliang Chen, Jiachui Zhou, and Zhaorong Yang. Pressure-induced reemergence of superconductivity in the topological kagome metal $\text{Cs}_3\text{V}_5\text{Sb}_5$. Phys. Rev. B 103, 224513 (2021).

doi: 10.1103/PhysRevB.103.224513

K.-Y. Chen, N.-N. Wang, Q.-W. Yin, Y.-H. Gu, K. Jiang, Z.-J. Tu, C.-S. Gong, Y. Uwamoto, J.-P. Sun, H.-C. Lei, J.-P. Hu, and J.-G. Cheng. Double superconducting dome and triple enhancement of $T_c$ in the kagome superconductor $\text{Cs}_3\text{V}_5\text{Sb}_5$ under high pressure. Phys. Rev. Lett. 126, 247001 (2021).

doi: 10.1103/PhysRevLett.126.247001

M. M. Denner, R. Thomale, and T. Neupert. Analysis of charge order in the kagome metal $\text{AV}_3\text{Sb}_5$ ($\text{A} = \text{K}, \text{Rb}, \text{Cs}$). Phys. Rev. Lett. 127, 217601 (2021).
23 T. Park, M. Ye, and L. Balents. Electronic instabilities of kagome metals: Saddle points and Landau theory. Phys. Rev. B 104, 035142 (2021).

doi: 10.1103/PhysRevB.104.035142

24 Y.-P. Lin and R. M. Nandkishore. Complex charge density waves at Van Hove singularity on hexagonal lattices: Haldane model phase diagram and potential realization in kagome metals AV3Sb5. Phys. Rev. B 104, 045122 (2021).

doi: 10.1103/PhysRevB.104.045122

25 X. Feng, K. Jiang, Z. Wang, and J. Hu. Chiral flux phase in the Kagome superconductor AV3Sb5. Science Bulletin 66, 1384 (2021).

doi: 10.1016/j.scib.2021.04.043

26 Z. Wang, Y.-X. Jiang, J.-X. Yin, Y. Li, G.-Y. Wang, H.-L. Huang, S. Shao, J. Liu, P. Zhu, N. Shumiya, M. S. Hossain, H. Liu, Y. Shi, J. Duan, X. Li, G. Chang, P. Dai, Z. Ye, G. Xu, Y. Wang, H. Zheng, J. Jia, M. Z. Hasan, and Y. Yao. Electronic nature of chiral charge order in the kagome superconductor CsV3Sb5. Phys. Rev. B 104, 075148 (2021).

doi: 10.1103/PhysRevB.104.075148

27 Z. Guguchia, C. Mielke III, D. Das, R. Gupta, J.-X. Yin, H. Liu, Q. Yin, M.H. Christensen, Z. Tu, C. Gong, N. Shumiya, T.S. Gamsakhurdashvili, M. Elender, Pengcheng Dai, A. Amato, Y. Shi, H.C. Lei, R.M. Fernandes, M.Z. Hasan, H. Luetkens, R. Khasanov, Tunable nodal kagome superconductivity in charge ordered AV3Sb5. arXiv:2202.07713 (2022).

28 L. Yu, C. Wang, Y. Zhang, M. Sander, S. Ni, Z. Lu, S. Ma, Z. Wang, Z. Zhao, H. Chen, K. Jiang, Y. Zhang, H. Yang, F. Zhou, X. Dong, S. L. Johnson, M. J. Graf, J. Hu, H.-J. Gao, and Z. Zhao. Evidence of a hidden flux phase in the topological kagome metal AV3Sb5.., arXiv:2107.10714 (2021).

29 Zhaoyang Shan, Pabitra K. Biswas, Sudeep K. Ghosh, T. Tula, Adrian D. Hillier, Devashibhai Adroja, Stephen Cotrell, Guang-Han Cao, Yi Liu, Xiaofeng Xu, Yu Song, Huiqiu Yuan, Michael Smidman. Muon-spin relaxation study of the layered kagome superconductor CsV3Sb5. arXiv:2203.05770 (2022).

30 Q. Wu, Z. X. Wang, Q. M. Liu, R. S. Li, S. X. Xu, Q. W. Yin, C. S. Gong, Z. J. Tu, H. C. Lei, T. Dong, N. L. Wang. The large static and pump-probe Kerr effect with two-fold rotation symmetry in Kagome metal AV3Sb5. arXiv:2110.11306 (2021).

31 R. Gupta, D. Das, C. H. Mielke III, Z. Guguchia, T. Shirola, C. Baines, M. Bartkowiak, H. Luetkens, R. Khasanov, Q. Yin, Z. Tu, C. Gong, and Hechang Lei. Microscopic evidence for anisotropic multigap superconductivity in the AV3Sb5 kagome superconductor. arXiv:2108.01574 (2021).

32 The Supplemental Information part contains information on the sample preparation and characterization procedure as well describes the details of muon-spin rotation/relaxation experiments.

33 R. Khasanov, H. Zhou, A. Amato, Z. Guguchia, E. Morenzoni, X. Dong, G. Zhang, and Z. Zhao. Proximity-induced superconductivity within the insulating (L0.84Fe0.16/OH layers in (L0.84Fe0.16/OHFe0.98Se. Phys. Rev. B 93, 224512 (2016).

doi: 10.1103/PhysRevB.93.224512

34 A. Amato, H. Luetkens, K. Sedlak, A. Stoykov, R. Scheuermann, M. Elender, A. Raselli, and D. Graf. The new versatile general purpose surface-muon instrument (GPS) based on silicon photomultipliers for µSR measurements on a continuous-wave beam. Rev. Sci. Instrum. 88, 093301 (2017).

doi: 10.1063/1.4986045

35 R. S. Hayano, Y. J. Uemura, J. Imazato, N. Nishida, T. Yamazaki, and R. Kubo. Zero-and low-field spin relaxation studied by positive muons. Phys. Rev. B 20, 850 (1979).

doi: 10.1103/PhysRevB.20.850

36 A. Suter and B. M. Wojek. Munifrit: A Free Platform-Independent Framework for µSR Data Analysis, Phys. Procedia 30, 69 (2012).

doi: 10.1016/j.phpro.2012.04.042

37 G. M. Luke, Y. Fudamoto, K. M. Kojima, M. I. Larkin, J. Merrin, B. Nachumi, Y. J. Uemura, Y. Maeno, Z. Q. Mao, Y. Mori, H. Nakamura, and M. Sigrist. Time-Reversal Symmetry Breaking Superconductivity in Sr2RuO4, Nature 394, 558-561 (1998).

doi: 10.1038/29038

38 A.D. Hillier, J. Quintanilla, B. Mazidian, J. F. Annett, and R. Cywinski. Nonunitary Triplet Pairing in the Cen-
trosymmetric Superconductor LaNiGa2, Phys. Rev. Lett. 109, 097001 (2012).

doi: 10.1103/PhysRevLett.109.097001

39 P. K. Biswas, H. Luetkens, T. Neupert, T. Stücer, C. Baines, G. Pascua, A.P. Schudyner, M. H. Fischer, J. Goryo, M. R. Lees, H. Maeter, F. Bru¨ckner, H.-H. Klauss, M. Nickelas, P. J. Baker, A. D. Hillier, M. Sigrist, A. Amato, and D. Johrendt. Evidence for superconductivity with broken time-reversal symmetry in locally noncentrosymmetric SrPtAs, Phys. Rev. B 87, 180503 (2013).

doi: 10.1103/PhysRevB.87.180503

40 V. Grinenko, S. Ghosh, R. Sarkar, J.-C. Orain, A. Nikitin, M. Elender, D. Das, Z. Guguchia, F. Bru¨ckner, M. E. Barber, J. Park, N. Kikugawa, D. A. Sokolov, J. C. Bobowski, T. Miyoshi, Y. Maeno, A. P. Mackenzie, H. Luetkens, C. W. Hicks, and H.-H. Klauss. Split superconducting and time-reversal symmetry-breaking transitions in Sr2RuO4 under stress. Nat. 17, 748 (2021).

doi: 10.1038/s41567-021-01182-7

41 J. E. Sonier, J. H. Brewer, R. F. Kieff, R. H. Heffner, K. F. Poon, S. L. Stubbs, G. D. Morris, R. I. Miller, W. N. Hardy, R. Liang, D. A. Bonn, C. E. Stronach, and N. J. Curro. Correlations between charge ordering and local magnetic fields in overdoped YBa2Cu3O6+x. Phys. Rev. B 66, 134501 (2002).

doi: 10.1103/PhysRevB.66.134501

42 H. Tan, Y. Liu, Z. Wang, and B. Yan. Charge Density Waves and Electronic Properties of Superconducting Kagome Metals. Phys. Rev. Lett. 127, 046401 (2021).

doi: 10.1103/PhysRevLett.127.046401

43 H. Li, T. T. Zhang, T. Yilmaz, Y. Y. Pai, C. E. Marinney, A. Said, Q. W. Yin, C. S. Gong, Z. J. Tu, E. Vescovo, C. S. Nelson, R. G. Moore, S. Murakami, H. C. Lei, H. N. Lee, B. J. Lawrie, and H. Miao. Observation of Unconventional Charge Density Wave without Acoustic Phonon Anomaly in Kagome Superconductors AV3Sb5 (A=Rb, Cs). Phys. Rev. X 11, 031050 (2021).

doi: 10.1103/PhysRevX.11.031050

44 Z. X. Wang, Q.Wu, Q.W. Yin, C. S. Gong, Z. J. Tu, T. Lin, Q. M. Liu, L. Y. Shi, S. J. Zhang, D. Wu, H. C. Lei, T. Dong, and N. L. Wang. Unconventional charge density wave and photoinduced lattice symmetry change in the kagome metal CsV3Sb5, probed by time-resolved spec-
troscopy. Phys. Rev. B 104, 165110 (2021).
doi: 10.1103/PhysRevB.104.165110

45 X. Zhou, Y. Li, X. Fan, J. Hao, Y. Dai, Z. Wang, Y. Yao, and H.-H. Wen. Origin of the Charge Density Wave in the Kagome Metal CsV₃Sb₅ as Revealed by Optical Spectroscopy. Phys. Rev. B 104, 041101 (2021).
doi: 10.1103/PhysRevB.104.L041101

46 R. Gupta, D. Das, C. Mielke III, E. Ritz, F. Hotz, Q. Yin, Z. Tu, C. Gong, H. Lei, T. Birol, R.M. Fernandes, Z. Guguchia, H. Luetkens, R. Khasanov. Two types of charge order in the superconducting kagome material CsV₃Sb₅. arXiv.2203.05055 (2022).

47 F. D. M. Haldane. Model for a Quantum Hall Effect without Landau Levels: Condensed-Matter Realization of the "Parity Anomaly". Phys. Rev. Lett. 61, 2015 (1988).
doi: 10.1103/PhysRevLett.61.2015

48 C. M. Varma. Non-Fermi-liquid states and pairing instability of a general model of copper oxide metals. Phys. Rev. B 55, 14554 (1997).
doi: 10.1103/PhysRevB.55.14554

49 X. Feng, Y. Zhang, K. Jiang, and J. Hu. Low-energy effective theory and symmetry classification of flux phases on the kagome lattice. Phys. Rev. B 104, 165136 (2021).
doi: 10.1103/PhysRevB.104.165136

50 J. Luo, Z. Zhao, Y. Z. Zhou, J. Yang, A. F. Fang, H. T. Yang, H. J. Gao, R. Zhou, Guo-qing Zheng. Star-of-David pattern charge density wave with additional modulation in the kagome superconductor CsV₃Sb₅ revealed by ⁵¹⁷V-NMR and ¹²¹/¹²³Sb-NQR. arXiv:2108.10263 (2021).

51 Y. Hu, X. Wu, B.R. Ortiz, X. Han, N.C. Plumb, S.D. Wilson, A.P. Schnyder, M. Shi. Coexistence of Tri-Hexagonal and Star-of-David Pattern in the Charge Density Wave of the Kagome Superconductor AV₃Sb₅. arXiv:2201.06477 (2022).

52 L. Nie, K. Sun, W. Ma, et. al., Charge-density-wave-driven electronic nematicity in a kagome superconductor. Nature (2022).
doi: 10.1038/s41586-022-04493-8

53 Y. Xiang, Q. Li, Y. Li, et. al., Twofold symmetry of c-axis resistivity in topological kagome superconductor CsV₃Sb₅ with in-plane rotating magnetic field. Nat Commun 12, 6727 (2021).
doi: 10.1038/s41467-021-27084-z

54 T. Neupert, M.M. Denner, J.-X. Yin, R. Thomale, and M.Z. Hasan. Charge order and superconductivity in kagome materials. Nature Physics 18, 137-143 (2022).
doi: 10.1038/s41567-021-01404-y

55 M. H. Christensen, T. Birol, B. M. Andersen, and R. M. Fernandes. Theory of the charge-density wave in AV₃Sb₅ kagome metals. Phys. Rev. B 104, 214513 (2021).
doi: 10.1103/PhysRevB.104.214513

56 Noah Ratcliff, Lily Hallett, Brenden R. Ortiz, Stephen D. Wilson, and John W. Harter. Coherent phonon spectroscopy and interlayer modulation of charge density wave order in the kagome metal CsV₃Sb₅. Phys. Rev. Materials 5, L111801 (2021).
doi: 10.1103/PhysRevMaterials.5.L111801

57 A. Subedi. Hexagonal-to-base-centered-orthorhombic 4Q charge density wave order in kagome metals KV₃Sb₅, RV₃Sb₅, and CsV₃Sb₅. Phys. Rev. Materials 6, 015001 (2022).
doi: 10.1103/PhysRevMaterials.6.015001

58 Shangfei Wu, Brenden R. Ortiz, Hengxin Tan, Stephen D. Wilson, Binghai Yan, Turan Birol, Girsh Blumberg. Charge density wave order in kagome metal AV₃Sb₅ (A= Cs, Rb, K). arXiv:2201.05188
I. CsV₃Sb₅ SINGLE CRYSTAL SAMPLES FOR µSR EXPERIMENTS

Figure 1 shows six single crystals of CsV₃Sb₅. The crystallographic c−axis is perpendicular to the flat surfaces of the crystals. The hexagonal symmetry of the crystals is obviously connected to the crystal shape. The red arrows represent directions of the crystallographic a−axis.

The Laue x-ray images of six CsV₃Sb₅ crystals from Fig. 1 are presented in Fig. 2. All images clearly demonstrate the hexagonal in-plane crystal structure of CsV₃Sb₅. The single crystallinity of the material and correspondence of the main crystal axes to the sample shape are visible.

II. AC SUSCEPTIBILITY RESPONSE OF CsV₃Sb₅ SINGLE-CRYSTALS

The superconducting response of CsV₃Sb₅ crystals was studied via ac susceptibility (ACS) experiments. CsV₃Sb₅ single crystals were placed inside the opened pressure cell (in our ACS experiments the pressure cell was not sealed and it was simply used as a sample container). The ACS setup and the pressure cell are described in Refs. 1, 2.

Figures 3 shows the ACS response of CsV₃Sb₅ single-crystals used in ZF-µSR [panel (a)] and TF-µSR [panel (b)] experiments as performed at GPS and HAL-9500 spectrometers, respectively. The transition temperatures T_c ≃ 2.62(3) and T_c ≃ 2.72(3) are determined from the crossing point of the linear fits of the ACS curve above and below T_c.

III. CSV₃SB₅ SINGLE-CRYSTALS ON µSR SAMPLE HOLDERS

Figure 4 illustrates the mounting of CsV₃Sb₅ single-crystals on the GPS sample holder. Panel (a) shows the a− and c−axis aligned CsV₃Sb₅ crystals attached to the sample mounting ring. The ring has 36 holes allowing to rotate it with a 10° step relative to the support plate.
FIG. 3: The temperature dependence of the ac susceptibility of CsV$_3$Sb$_5$ single-crystals used in ZF-μSR [panel (a)] and TF-μSR [panel (b)] experiments, as performed at the GPS and HAL-9500 spectrometers, respectively. The transition temperature $T_c$ is obtained from the crossing point of the linearly extrapolated ACS curves in the vicinity of $T_c$.

Panels (b), (c), and (d) correspond to the positioning of the samples at angle $\alpha = 0^\circ$, $30^\circ$, and $60^\circ$, respectively.

Figure 4 (a) shows CsV$_3$Sb$_5$ single-crystals mounted on top of the HAL-9500 sample holder (made of 99.999% pure silver). Panel (b) demonstrates the sample holder attached to the cold finger of the continuous-flow He4 cryostat.

FIG. 4: (a) CsV$_3$Sb$_5$ single-crystals mounted on the holding ring of the GPS sample holder. (b), (c), and (d) the sample ring attached to the sample holder and turned at the angle $\alpha = 0^\circ$ [panel (a)], $30^\circ$ [panel (b)] and $60^\circ$ [panel (c)], respectively.

FIG. 5: (a) CsV$_3$Sb$_5$ single-crystals mounted on the HAL-9500 sample holder. (b) The sample holder with crystals attached to the cold finger of the continuous-flow He4 cryostat.

IV. THE BACKGROUND ESTIMATE

In μSR experiments, part of the muons may not hit the sample and stop in the sample holder, cryostat windows, cryostat walls, etc. These muons contribute in the background response, which must be known prior to performing the data analysis.

A. The background contribution in GPS experiments

In experiments performed at the GPS spectrometer, the samples (CsV$_3$Sb$_5$ single-crystals) were mounted on the holder shown in Fig. 4. The background was estimated from measurements made in the superconducting state. The external magnetic field $B_{ext} = 10$ mT was applied parallel to the crystallographic c−axis at $T = 5$ K, i.e. above the superconducting transition temperature $T_c \approx 2.6$ K [see Fig. 3 (a)]. The angles $\alpha$ and $\beta$ i.e. the angle between the crystallographic c−axis and the in-plane component of the muon-spin polarization $P_{\mu}^{||}$ and the angle between the muon momentum $p_{\mu}$ and the initial muon-spin polarization $P_{\mu}$, we kept at $\alpha = 30^\circ$ and $\beta = 45^\circ$, respectively.

The Fourier transforms of TF-μSR time spectra, representing the internal field distribution $P(B)$, are shown in Fig. 6. The panel (a) corresponds to $P(B)$ obtained...
In high-field experiments performed at the HAL-9500 spectrometer under an applied field of \( B_{\text{ext}} = 8 \) T, a polarizations of the sample and the background component, respectively. The background contribution was approximated by a cosine decay function:

\[
P_{\text{BG}}(t) = \exp(-\sigma_{\text{BG}}^2 t^2 / 2) \cos(\gamma_{\mu} B_{\text{ext}} t + \phi).
\]  (2)

Here \( \gamma_{\mu} = 2\pi 135.5 \text{ MHz/T} \) is the muon gyromagnetic ratio, \( \sigma_{\text{BG}} \) is the Gaussian relaxation rate and \( \phi \) is the initial phase of the muon-spin ensemble. In order to account for the asymmetric field distribution \( P(B) \) caused by formation of the flux-line lattice (FLL) in the superconducting state, the sample contribution was described using the Skewed Gaussian (SKG) distribution function:

\[
P_S(t) = \text{SKG}(t) \cos(\gamma_{\mu} B_0 t + \phi).
\]  (3)

Here \( B_0 \) is the field corresponding to the maximum of \( P(B) \) distribution of SKG\( (t) \) function.\(^3\) The fit of Eq. 1 with the sample and the background components described by Eqs. 2 and 3 results in \( A_{0, \text{BG}} / A_0 \approx 0.064 \) and 0.045 for the experimental data presented in Figs. 6 (c) and (d), respectively. As an average value, \( \sim 5\% \) background contribution was considered for experiments performed at the GPS spectrometer.

The background contribution might be visualized by comparing the 'total' \( P(B) \) distributions with the field distributions caused by formation of the FLL inside the superconducting \( \text{CsV}_3\text{Sb}_5 \) [see the corresponding red and blue lines at insets to Figs. 6 (c) and (d)]. The difference, as is marked by pink areas, represent the background contribution.

**B. Background for experiments performed at the HAL-9500 spectrometer**

In high-field experiments performed at the HAL-9500 spectrometer under an applied field of \( B_{\text{ext}} = 8 \) T, a...
FIG. 8: (a)–(c) The asymmetry spectra collected for the $P^{||ab}(t)$ set of experiments at $T = 5$ K, $\alpha = 30^\circ$ and $\beta = 60^\circ$. The red and black curves correspond to the response of positron detectors staying in-phase and out-of-phase with $P^{||ab}$. (d)–(f) The asymmetry spectra collected for the $P^{||ab}(t)$ set of experiments at $T = 5$ K, $\alpha = 30^\circ$ and $\beta = 5^\circ$. The solid lines in panels (a)–(f) are fits of Eq. 1 from the main text to the experimental data. The fits were made with both – the Gaussian Kubo-Toyabe $\sigma_{\text{GKT}}$ and the exponential $\lambda$ – relaxations remaining free [panels (a) and (d)]; with $\sigma_{\text{GKT}}$ and $\lambda$ fixed to zero [panels (b) and (e)]; and with $\sigma_{\text{GKT}} = 0$ and $\lambda$ free [panels (c) and (f)].

two-component signal was observed within the full range of temperatures. Figure 7 shows the field distribution $P(B)$ measured at $T = 20$ K. The solid line corresponds to the two component fit by means of Eq. 1. The background and the sample component were described by the aforementioned Eq. 2 and by Eq. 2 from the main text, respectively. The background contribution is visualized as a difference between the two-component fit and the sample contribution (see the pink area at the inset to Fig. 7).

The width and the position of the background signal were found to be temperature independent, as expected, and they were kept fixed during the analysis ($A_{0,\text{BG}}/A_0 \simeq 0.2$, $\sigma_{\text{BG}} \simeq 0.16$ $\mu$s$^{-1}$).

V. THE PRESENCE OF NUCLEAR AND ELECTRONIC COMPONENTS IN $\mu$SR RESPONSE

Validity of the data analysis procedure considering the presence of both – the electronic and the nuclear components – was checked for the set of ZF-$\mu$SR data collected at $T = 5$ K for two different components of the muon-spin polarization [$P^{||ab}(t)$ and $P^{||c}(t)$]. The red and black curves correspond to the data collected at positron detectors staying in-phase ($0^\circ$) and out-of-phase ($180^\circ$) to the corresponding initial component of the muon-spin polarization. Three different fit types of Eq. 1 from the main text to the experimental data were performed:

1. Both, $\lambda$ and $\sigma_{\text{GKT}}$, remain free, i.e., stay fitted [panels (a) and (d)]
2. $\sigma_{\text{GKT}}$ stays free and $\lambda$ is fixed to '0' [panels (b) and (e)].
3. $\lambda$ remains free and $\sigma_{\text{GKT}} = 0$ [panels (c) and (f)].

The results of the fit of Eq. 1 from the main text to the data are presented in Fig. 8 by solid lines.

| Fit type | $\lambda$ ($\mu$s$^{-1}$) | $\sigma_{\text{GKT}}$ ($\mu$s$^{-1}$) | $\chi^2_{\text{norm}}$ |
|----------|-----------------|-----------------|----------------|
| $P^{||ab}$ | free $\lambda$ and $\sigma_{\text{GKT}}$ | 0.0118(40) | 0.2231(22) | 1.210 |
| | $\lambda = 0$ and free $\sigma_{\text{GKT}}$ | 0 | 0.2281(13) | 1.262 |
| | free $\lambda$ and $\sigma_{\text{GKT}} = 0$ | 0.1857(25) | 0 | 12.31 |
| $P^{||c}$ | free $\lambda$ and $\sigma_{\text{GKT}}$ | 0.0453(47) | 0.1593(42) | 1.190 |
| | $\lambda = 0$ and free $\sigma_{\text{GKT}}$ | 0 | 0.1900(16) | 1.968 |
| | free $\lambda$ and $\sigma_{\text{GKT}} = 0$ | 0.1204(25) | 0 | 3.349 |

The fitted relaxation rates are summarised in Table I.
The goodness of each fit type was evaluated by calculating the values of the normalized $\chi^2$ (the sum of the mean squared deviations divided by the number of degrees of freedom). Note that, in their normalized form, $\chi^2$ takes into account the decreased number of the fit parameters for cases when one of the relaxation rates (\(\lambda\) or \(\sigma_{\text{GKT}}\)) was fixed to zero.

The results presented in Fig. 8 and Table I imply that the experimental data require the presence of both – the exponential \(\lambda\) and the Gaussian Kubo-Toyabe \(\sigma_{\text{GKT}}\) – relaxation components. Exclusion of either \(\lambda\) or \(\sigma_{\text{GKT}}\) leads to substantial increase of $\chi^2$. This agrees, therefore, with the results of Ref. 4, where the ZF-$\mu$SR data of KV$_3$Sb$_5$ representative of AV$_3$Sb$_5$ family were described by using both – \(\sigma_{\text{GKT}}\) and \(\lambda\) relaxation components. A similar approach was used in the majority of other TRSB $\mu$SR studies as reported e.g. in Refs. 5–14. Our results would also suggest that the analysis of CsV$_3$Sb$_5$ and KV$_3$Sb$_5$ ZF-$\mu$SR data from Refs. 15 and 16, where only the Gaussian Kubo-Toyabe term was taken into account, need to be reconsidered.

VI. COMPARISON BETWEEN THE ZERO-FIELD AND LONGITUDINAL-FIELD DATA

Both static and fluctuating internal magnetic fields can have an effect on the exponential muon-spin polarization relaxation rate as is measured in ZF experiments. In order to differentiate between static and fluctuating internal magnetic fields, the longitudinal-field (LF) experiments with the externally applied magnetic field \(B_{\text{ext}} = 3\) mT applied along the \(P_{\parallel ab}\) component of the muon-spin polarization were conducted. The results reveal that the Gaussian Kubo-Toyabe component observed in ZF-$\mu$SR is absent, as expected when the muon-spins are decoupled from the local field, while the exponential relaxation rate \(\lambda\) remains at a certain non-zero value.

Figure 9 compares the exponential relaxation rates as they observed in ZF and LF set of $\mu$SR experiments. From the data presented in Fig. 9 two important points emerge:

(i) The LF relaxation points stay systematically lower the ZF ones. This suggests that 3 mT magnetic field leads to partial, but not complete, decoupling of muon-spins from internal fields.

(ii) The LF relaxation rate decreases nearly linearly within the full temperature range studied, \(i.e.\) from \(\approx 3\) up to \(\approx 300\) K. This imply that the dynamical component on the exponential relaxation is indeed present. However it is not related to the appearance of the charge density order below \(T_{\text{CDW}} \approx 95\) K, as well as to an additional enhancement of the ZF relaxation below \(T^* \approx 30\) K.

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1 R. Khasanov, Z. Guguchia, A. Maisuradze, D. Andreica, M. Elender, A. Raselli, Z. Shermanidi, T. Goko, F. Neutech, E. Morenzoni, and A. Amato, High pressure research using muons at the Paul Scherrer Institute, High Pressure Research 36, 140-166 (2016).
doi: 10.1080/08957959.2016.1173690
Z. Shermadini, R. Khasanov, M. Elender, G. Simutis, Z. Guguchia, K.V. Kamenev, and A. Amato, A low-background piston-cylinder type hybrid high pressure cell for muon-spin rotation/relaxation experiments, High Pressure Research 37, 449-464 (2017). doi: 10.1080/08957955.2017.1373773

A. Suter, Preprint at http://imu.web.psi.ch/docu/LEM Memo/skewedGaussian/skewedGaussian.pdf (2008).

T. Shang, M. Smidman, S. K. Ghosh, C. Baines, L. J. Merrin, B. Nachumi, Y. J. Uemura, Y. Maeno, A. P. Mackenzie, H. Luetkens, C. W. Hicks, and H.-H. Klauss. Split superconducting and time-reversal symmetry-breaking transitions in Sr$\text{RuO}_4$ under stress. Nat. Phys. 17, 748 (2021). doi: 10.1038/s41467-021-01182-7

V. Grinenko, R. Sarkar, K. Kihou, C. H. Lee, I. Morozov, S. Aswartham, B. Büchner, P. Chekhonin, W. Skrotzki, K. Kenkov, R. Hühne, K. Nielsch, S. L. Drechsler, V. L. Vadinov, M. A. Silaev, P. A. Volkov, I. Eremin, H. Luetkens, and H.-H. Klauss. Superconductivity with broken time-reversal symmetry inside a superconducting s-wave state. Nat. Phys. 16, 789 (2020). doi: 10.1038/s41567-020-0886-9

V. Grinenko, S. Ghosh, R. Sarkar, J.-C. Orain, A. Nikitin, M. Elender, D. Das, Z. Guguchia, F. Brückner, M. E. Barber, J. Park, N. Kikugawa, D. A. Sokolov, J. C. Bobowski, T. Miyoshi, Y. Maeno, A. P. Mackenzie, H. Luetkens, C. W. Hicks, and H.-H. Klauss. Split superconducting and time-reversal symmetry-breaking transitions in Sr$\text{RuO}_4$ under stress. Nat. Phys. 17, 748 (2021). doi: 10.1038/s41467-021-01182-7

E. M. Kenney, B. R. Ortiz, C. Wang, S. D. Wilson, and M. J. Graf. Absence of local moments in the kagome metal KV$_3$Sb$_5$ as determined by muon spin spectroscopy. J. Phys.: Condens. Matter 33 235801 (2021). doi: 10.1088/1361-646X/abe8f9