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High Resolution Polar Kerr Effect Studies of CsV_3Sb_5: Tests for Time Reversal Symmetry Breaking Below the Charge Order Transition

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We report high resolution polar Kerr effect measurements on CsV_3Sb_5 single crystals in search for signatures of spontaneous time reversal symmetry breaking below the charge order transition at $T^* \approx 94$ K. Utilizing two different versions of zero-area loop Sagnac interferometers operating at 1550 nm wavelength, each with the fundamental attribute that without a time reversal symmetry breaking sample at its path, the interferometer is perfectly reciprocal, we find no observable Kerr effect to within the noise floor limit of the apparatus at 30 nanoradians. Simultaneous coherent reflection ratio measurements confirm the sharpness of the charge order transition in the same optical volume as the Kerr measurements. At finite magnetic field we observe a sharp onset of a diamagnetic shift in the Kerr signal at T^*, which persists down to the lowest temperature without change in trend. Since 1550 nm is an energy that was shown to capture all features of the optical properties of the material that interact with the charge order transition, we are led to conclude that it is highly unlikely that time reversal symmetry is broken in the charge ordered state in CsV_3Sb_5 .

Symmetry-breaking is the phenomenon where an infinitesimal perturbation can cause the system to break the underlying symmetry of the Hamiltonian. And it is a cornerstone concept in the understanding and manipulation of quantum materials. The state of a system can also be manipulated without explicit symmetry-breaking by controlling topological aspects of the material in momentum space, thereby regulating the electronic bandstructure. When strong electron correlations dominate, the system is often observed to be close to multiple competing ordered phases with similar energies. For example, the appearance of charge order may trigger competition with an emerging new electronic state, which in turn leads to the phenomenon of "intertwined order" [1].

A particularly interesting material system in that respect are the quasi-two-dimensional kagomé compounds AV_3Sb_5 (A=K, Rb and Cs) [2]. These are layered materials with ideal kagomé lattice of V ions coordinated by Sb crystallizing in the P6/mmm space group. Cooling these materials below a temperature T^*, the kagomé lattice distorts, undergoing a charge-order transition ($T^* \approx$ 80 K, 110 K and 94 K for K, Rb and Cs respectively) [3–5], which was shown to be associated with a CDW superstructure modulation displaying chiral anisotropy [6–8]. Further investigation of the CDW transitions using hard-X-ray scattering revealed that the observed superstructure has in fact a three-dimensional (3D) nature with either $2 \times 2 \times 4$ or $2 \times 2 \times 2$ superstructure depending on conditions of crystal growth, imposed strain and sample's thermal history [9–13]. Intertwined order was initially identified with the discovery of superconductivity in this material system with $T_c \approx 2.8$ K for CsV_3Sb_5 [3], where the charge-order transition was first suspected to be wholly electronic in origin. However, detailed nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) measurements on CsV_3Sb_5 single crystals revealed an orbital ordering at $T^* \approx 94$ K clearly induced by a first order structural transition. This is accompanied by electronic charge order that appears to grow gradually below T^* as a secondary (intertwined) order [14]. In fact, a first order characteristic of the transition can be further deduced from the anomalous peak of the specific heat [3, 12, 15] and the abrupt large diamagnetic shift of the magnetic susceptibility for magnetic field aligned with the c-axis [3, 15]. The absence of such diamagnetic shift at T^* for magnetic field in the ab plane for KV_3Sb_5 (see "Extended Data," Fig. 3 in [16]) and the much reduced effect for CsV_3Sb_5 [17] further support the NMR/NQR findings.

Focusing on the transitions at T^*, a key issue of a possible co-occurrence of time-reversal symmetry breaking (TRSB) has emerged through claims of observation of

anomalous Hall effect (AHE)[15, 18, 19], changes in muon spin relaxation rate below T^* [16, 20, 21], and magneto-optic Kerr effects [22–24]. From a theoretical standpoint, a CDW formation in kagome metals can appear through both electronically and phonon-mediated mechanisms, or a cooperative version thereof. For CsV_3Sb_5 , both electronic and phonon fluctuations seems to be relevant due to the joint presence of van Hove singularities nearby the Fermi level and prominent breathing-phonon modes [25–28]. The possible appearance of orbital currents has suggested the possibility of a TRSB state below T^*, along with an enhanced propensity to nematicity due to multiple CDW nesting vectors [26, 29]. Both TRSB and point group symmetry breaking via nematicity are secondary to the translation symmetry breaking and dependent on the precise microscopic setting involving temperature, disorder, and interaction profile.

In this paper we aim to closely examine possible TRSB in high quality crystals of CsV_3Sb_5 via high resolution measurements of the polar Kerr effect. To substantiate our findings, we use crystals grown in two different laboratories (MPI-Dresden and UC Santa Barbara), all showing salient attributes of the charge order transition at $T^* \approx 94$ K. Further we use two different zero-area-loop Sagnac interferometers (ZALSI) [30, 31] operating at a wavelength of 1550 nm, one at Stanford University and one at UC Irvine to measure Kerr rotation through the CDW transition, both in zero and in finite magnetic fields. While we clearly observe the abrupt onset of optical birefringence and/or dichroism associated with the structural transition, we see no evidence for a spontaneous Kerr effect below T^* within the volume of the same optical beam to a measurement limit of ± 30 nanoradians, neither in zero-field cool, nor after training with a magnetic field up to 0.34 T. Measurements in an applied magnetic field reveal a diamagnetic Kerr shift, which onsets abruptly below T^* similar to the magnetic susceptibility [3], but unlike the susceptibility, it stays diamagnetic down to low temperatures.

Our results stand in stark contrast to previously published optical measurements claiming finite spontaneous Kerr effects. Two experiments [22, 23] performed at 800 nm wavelength measured rotation of linearly polarized light at oblique reflection angle, while the third [24] used a similar apparatus to our ZALSI at 1550 nm. We discuss the differences between our measurement and reports [22, 23] in detail below. As for the third report [24], we speculate that a series of uncontrolled subtractions of data may have been the source of a false positive result.

In this study we use high quality and well-characterized single crystals of CsV_3Sb_5 obtained from two different sources. Samples grown at UCSB (dubbed sample 2) followed the previously published procedure found in [3] and related publications, while samples grown at MPI-Dresden (dubbed sample 1) followed the previously published procedure found in [32] and related publications. The crystal structure of CsV_3Sb_5 and photos of two example crystals from the two sources are presented in Fig. 1. Specific heat data obtained on these crystals show the "standard" sharp first–order–like transition peak at 93.5 K as is shown in Fig. 2. This charge density transition is also marked by the kink in DC resistance and the pronounced peak in its first derivative dR/dT as shown in Supplemetary Figure [33]. Moreover, we observe two additional features in the dR/dT curve. There is a dispersive line shape close to the temperatures where a kink was observed in NMR $1/T_1T$ [14] and where the electronic magneto-chiral anisotropy (eMChA) was observed in nonlinear transport [34], suggesting an additional phase transition. Also there is a small bump in resistance derivative happening at temperatures below T^* which could be related to either onset of CDW along c direction or to formation of $4a_0$ stripe order [35]. Importantly, we also detect onset of charge order simultaneously with Kerr effect measurements within the same optical volume through reflectivity and coherent reflection ratio measurements.

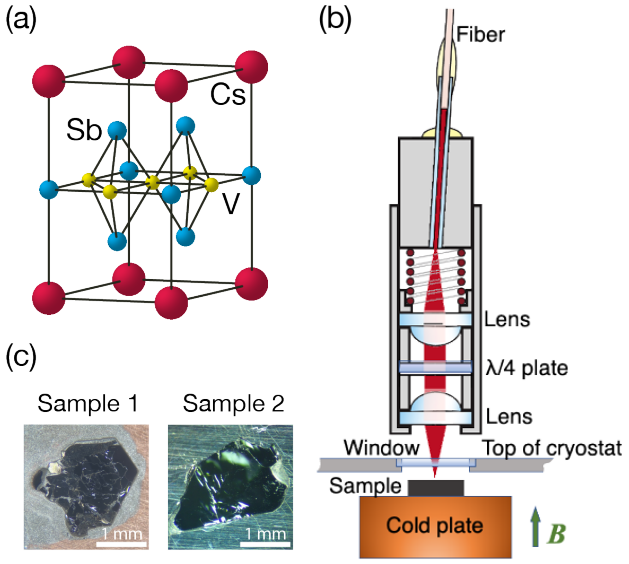

FIG. 1: (a) Crystal structure of CsV_3Sb_5 . (b) Photo of "sample 1" from Dresden, measured at Stanford and "sample 2" from UCSB, measured at UC Irvine. (c) Schematic cross-section of probe end optics assembly and sample (not to scale).

High resolution Polar Kerr effect measurements presented in this paper were performed using zero-area-loop Sagnac interferometers (ZALSI) first introduced by Xia *et al.* [30] and successfully used to probe different systems [36–41]. By construction the ZALSI is inherently reciprocal and thus by its symmetry it can distinguish between true TRSB Kerr effect and optical activity when reflected from a chiral medium[42]. The light beam of ZALSI is fo-

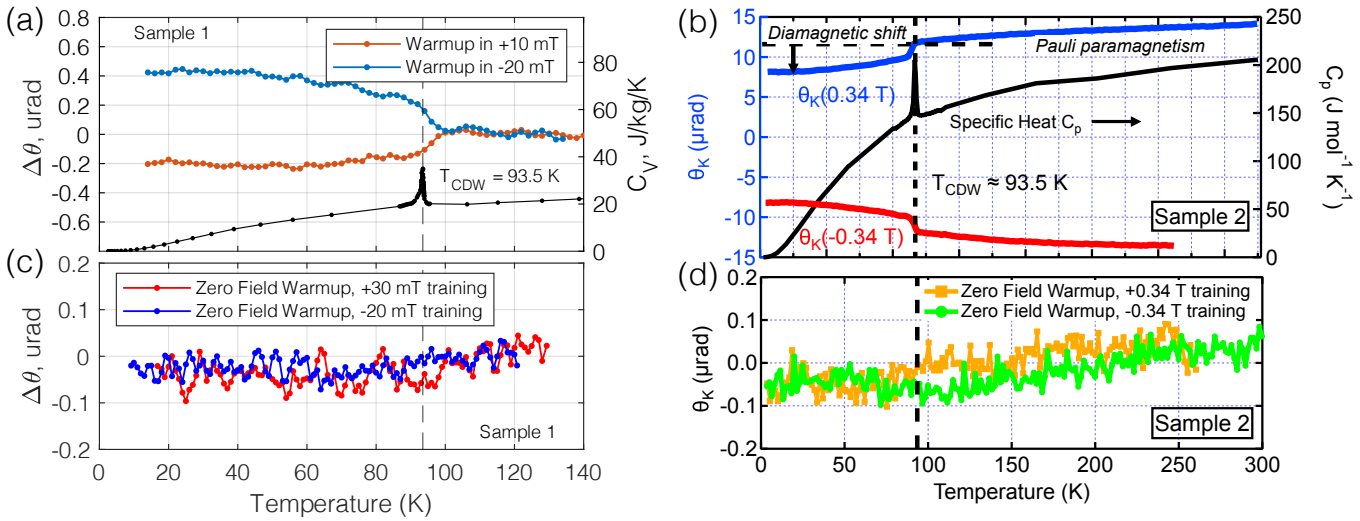

FIG. 2: **Kerr signal in CsV_3Sb_5.** (a) Sample 1 measured at Stanford: Relative Kerr angle $\Delta\theta = \theta - \theta(120K)$ (left axes) in external magnetic fields of 20 mT directed along $\pm\hat{z}$, showing the onset of diamagnetic shift when the specific heat C_V (right axes) develops a peak at $T^* \approx 94$ K. (b) Sample 2 measured at Irvine: Absolute Kerr signal θ_K (left axes) during ± 0.34 T cooldowns showing the same onset of diamagnetic shift that coincides with the anomalous peak in specific heat C_p (right axes). (c) Sample 1: Kerr angle $\Delta\theta$ measured during zero field warmups after +30 mT and -20 mT trainings. (d) Sample 2: Kerr signal θ_K measured during zero field warmups after ± 0.34 T trainings. There is no discernible spontaneous Kerr effect below T^* to within 30 nrad in either samples.

cused onto an optically flat region that can be hundreds of microns in size. One such region is illustrated in the scanning images of reflected power P_0 on sample 2 at $3.1K$ provided in supplementary materials [33]. In a magnetic field of -0.34 T the Kerr signal is uniform across this flat region. The temperature dependence of the in-field Kerr signals at fixed spots are shown in Fig. 2A for sample 1 measured at Stanford, and in Fig. 2B for sample 2 measured at Irvine, alongside the measured specific heat. In both cases, there is a clear and relatively abrupt onset of diamagnetic shift that coincides with the anomalous peak in the specific heat at $T_{\rm CDW}$, confirming the CDW transition in the probed optical volume.

The main result of this work is the test for spontaneous Kerr effect in CsV_3Sb_5 . Kerr signal was recorded during zero field warmup after removal of a "training" magnetic field applied during cooldown. Results from the two laboratories are presented in Fig. 2C and Fig. 2D respectively, showing no discernible Kerr effect below T^* to within 50 nrad. This uncertainty is a factor of $\sim$ 1000 smaller than the recent MOKE measurements [22–24]. Similarly, no spontaneous Kerr signal was found during zero field cooldowns and subsequent zero field warmups. The absence of spontaneous Kerr signal is also universally found across the sample surface, which can be seen in the scanning Kerr signal image presented in the supplementary materials [33] taken on sample 2 at $3.1K$ after removal of the -0.34 T magnetic field.

Our main result raises doubts to whether time reversal symmetry is indeed broken below T^* in CsV_3Sb_5 . On the other hand, measurements at a finite magnetic field oriented along the c-axis show a clear and relatively abrupt diamagnetic shift in the Kerr effect, which near T^* behaves similarly to the in-field magnetic susceptibility measurements [3, 8, 15]. However, the detected average (DC) optical power (P_0) and the coherent reflection ratio (P_2/P_0) (see supplementary material [33]) measurements in either zero field or in-field, which are taken simultaneously with the Kerr measurements within the same optical volume, consistently show an abrupt transition at T^* with a width of $\sim$ 2 to 3 K, irrespective of the applied fields (similar result is reported in [43, 44]). Since TRSB effect is expected to be secondary to any structural or CDW transitions, and grow gradually below T^* [26], it is unlikely that the observed abrupt changes in the differential reflectivity are associated with such a TRSB effect. A similar scenario was previously observed in the Cuprate superconductors where a first-order structural transition from an orthorhombic to tetragonal phase at T_s (so called LTO to LTT transition) in $La_{2-x}Ba_xCuO_4$ [45] was accompanied by a charge-order transition with an order parameter that evolved gradually below T_s.

The bulk magnetic susceptibility of CsV_3Sb_5 has been previously shown to develop a paramagnetic response below $\sim$ 50 K [3, 8, 15], presumably due to bulk impurities, overwhelming the initial diamagnetic drop below T^*. The Sagnac interferometers probe only the freshly cleaved surface region within a small volume defined by

the beam diameter $d = 10$ μm and the optical penetration depth $\delta \sim 20$ nm, and thus are less susceptible to these bulk impurities, particularly as we typically use a lower magnetic field. Hence, the change in the Kerr signal below T^* stays diamagnetic and is almost temperature independent as seen in the relative Kerr angle $\Delta\theta = \theta - \theta(120K)$ for sample 1 in Fig. 2A and in the absolute Kerr signal θ_K for sample 2 in Fig. 2B.

The susceptibility measurements on CsV_3Sb_5 further show that the diamagnetic drop with the field along the ab plane is almost 5 times weaker than if the field points in the c-direction, where earlier results on KV_3Sb_5 show no diamagnetic shift for the field along the ab-plane [3]. These observations indicate that the diamagnetic response originates from in-plane effect, presumably orbital effects such as non-interacting loop currents as suggested by the NMR/NQR experiments [14]. In contrast, the paramagnetic response observed in magnetic susceptibility is isotropic with respect to the applied magnetic field and remains on the scale of the Pauli paramagnetism above T^*. Hence, we conclude that we have no evidence for spontaneous ferromagnetic orderings of loop currents. However, our results still allow for an antiferromagnetic ordering of loop currents.

Next we provide our speculations on why results reported here are in contrast to other optical measurements that attempt to measure Kerr effect in the AV_3Sb_5 systems [22, 23]. First of all, we note that while the ZALSI is designed to only detect TRSB effects in the sample as an excess signal beyond the natural reciprocity of the apparatus, other techniques may need to employ less controlled means to scrutinize true TRSB effects. Secondly, we note that measurements [22, 23] have been made at oblique incident angle, while we have measured light reflected at normal incidence. It is known that, in an optically active material, the polarization of reflected light at oblique incidence will always rotate with respect to the initial state of polarization irrespective of orientation between initial polarization and principal axes of the crystal [46, 47]. Furthermore, the statement that no finite optical rotation is possible for reflected light at normal incidence is true for polar Kerr effect, but does not apply to circular dichroism, which originates from different optical penetration depths for the two circularly polarized components. Thirdly, if circular dichroism is detected at normal incidence, it is not a proof for TRSB. In fact, in most cases it will indicate circular birefringence that respects time reversal symmetry. A notable example is reflection from the surface of tellurium, where large rotary power and circular dichroism were previously measured [48–50], but no Kerr effect was detected using our ZALSI [51]. Lastly, concerning the difference in photon wavelength used in our system and in [22, 23], we turn to the optical properties of CsV_3Sb_5 [43, 44, 52] and note that our wavelength is close to the Lorenz peak in optical conductivity centered around 0.6 μm^{-1} wavenumbers (dubbed L_1 in [43]), which is enhanced by CDW. Hence, we would expect that our wavelength probes exactly the electronic states responsible for charge order formation, unlike the $\sim$ 800 nm wavelength which is further away from the CDW energy scale. Thus, we conclude that unless there is a special reason for Kerr rotation to zero at 1550 nm, which is unlikely in a good metal, our wavelength should provide a more sensitive probe compared to the one used in [22, 23].

The above discussion suggests that at least within the technical aspect, those other optical measurements fall short in comparison to the ZALSI, which inherently measures only non-reciprocal effects, thus returning a zero signal if time reversal symmetry is not broken. On the materials side we note that if the CDW is chiral along the c-axis of the material, mirror symmetries are broken as was previously observed in STM [6–8], and circular dichroism is allowed. Moreover, a finite minimum value of polarization rotation will be present with magnitude that depends on the pitch of the chiral CDW, which depends on the initial symmetry of the unbroken state, as well as the optical penetration depth at the probing photon energy. These effects may then be incorrectly interpreted as signatures of TRSB. From the technical point of view we also note that using linear polarization to probe complex materials requires perfect alignments of the optical components with respect to the plane of incidence. For example, if rotation of the incident light polarization is performed using a perfect half-wave plate (HWP), which is tilted by a small angle with respect to the propagation axis, even a perfect metallic mirror will show an apparent polarization rotation [53]. For CsV_3Sb_5 parameters, a 5 degrees error in the HWP alignment yields a ± 1 millirad error. Commercial zero-order HWPs have a typical retardance error of 1% even for normal incidence, yielding further errors.

Finally we briefly comment on the reported onset of anomalous Hall effect [15, 18, 19] and change in muon spin relaxation below T^* [16, 20, 21]. Indeed, the anomalous Hall effect is a direct measure of ferromagnetic order. However, in general it implies a finite value at zero applied magnetic field. Yet, all reports of AHE in CsV_3Sb_5 show no Hall resistance at zero magnetic field. In that respect this AHE is not strictly related to spontaneous TRSB effect and may come from non-TRSB chiral effects [54] or change in carrier density and mobility due to charge order. Another set of experiments which are at odds with our results involve μ-Sr measurements, and in particular the study in [16] where a TRSB is inferred from a rather small change through a sharp onset in muons relaxation below T^* at zero field, followed by an almost temperature independent trend at low temperatures. Indeed, as a purely magnetic probe, μSr is not directly sensitive to charge order and thus a change in relaxation might be interpreted as evidence for TRSB in the electronic system. However, μSr does sense the pres-

ence of charge order via the magnetic dipolar coupling of the muons with the host nuclei at zero or low magnetic fields (see e.g. [55]). In particular, since the zero and low field effect in the μSr studies is abrupt and follows in trend the structural transition [14], it is likely to induce a change in the muons' preferred "rest positions" above and below T^*, which will show up as change in relaxation rates. While an all-electronic spontaneous TRSB buildup mechanism for the CDW [26] is in principle not inconsistent with the μSr data deeper in the CDW phase, the abruptness in the μSr response at the transition cannot solely be reconciled from this angle, and suggests a pivotal role of phonon-mediated CDW formation related to a structural first order transition. Furthermore, the in-field enhancement of the signal in magnetic field seems to follow the NMR results, which again cannot directly point to a spontaneous TRSB.

In conclusion, we have used high resolution polar Kerr effect measurements to scrutinize previously reported submissions [22–24] that time reversal symmetry is spontaneously broken below the charge order transition a in CsV_3Sb_5 (marked as T^*). For these studies we used two different variation of a zero-area loop Sagnac interferometer operating at 1550 nm wavelength, each with the fundamental attribute that without a time reversal symmetry breaking sample at its path, the interferometer is perfectly reciprocal and thus yield a zero Kerr signal at the detector to within its noise floor limit of ~ 30 nanoradians. We then show that high quality CsV_3Sb_5 single crystals obtained from two different crystal-growth laboratories show no resolvable Kerr signal at zero magnetic field to within the instruments noise floor level, whether the sample was cooled at zero field, or was trained through T^* at a finite field and then measured at zero field upon warming up. Concurrent measurements of the coherent reflection ratio show a sharp transition within the same optical volume where Kerr effect was measured, thus supporting the robustness of our measurements. At finite magnetic field we observe a sharp diamagnetic transition at T^*, similar to the transition observed in magnetic susceptibility, except that the Kerr data remain diamagnetic down to low temperatures without change in trend. Since 1550 nm is an energy that was shown to capture all features of the optical properties of the material that interact with the charge order transition, we are led to conclude that it is highly unlikely that time reversal symmetry is broken in the charge ordered state in CsV_3Sb_5 .

Note added during the review process, we've learned that recent measurement [56] demonstrated that rotation of linear polarization in CsV_3Sb_5 has isotropic component θ_C even at normal incidence that is comparable in size to those found at 800 nm [22, 23]. Moreover, it was found that θ_C does not respond to applied magnetic field and changes sign and magnitude from sample to sample. These suggest that the polarization rotations reported in [22, 23] may not be related to MOKE.

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