

Supplemental Information for “Time-reversal symmetry breaking in charge density wave of CsV_3Sb_5 detected by polar Kerr effect”

Yajian Hu^{1*}, Soichiro Yamane¹, Giordano Mattoni^{1,2}, Kanae Yada¹, Keito Obata¹, Yongkai Li^{3,4,5}, Yugui Yao^{3,4}, Zhiwei Wang^{3,4,5}, Jingyuan Wang⁶, Camron Farhang⁶, Jing Xia⁶, Yoshiteru Maeno^{1,2}, Shingo Yonezawa^{1†}

¹*Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan*

²*Toyota Riken-Kyoto University Research Center (TRiKUC), Kyoto University, Kyoto 606-8501, Japan*

³*Centre for Quantum Physics, Key Laboratory of Advanced Optoelectronic Quantum Architecture and Measurement (MOE), School of Physics, Beijing Institute of Technology, Beijing 100081, P. R. China*

⁴*Beijing Key Lab of Nanophotonics and Ultrafine Optoelectronic Systems, Beijing Institute of Technology, Beijing 100081, P. R. China*

⁵*Material Science Center, Yangtze Delta Region Academy of Beijing Institute of Technology, Jiaxing 314011, P. R. China*

⁶*Department of Physics and Astronomy, University of California, Irvine, California 92697, USA*

*e-mail: hu.yajian.78e@st.kyoto-u.ac.jp †e-mail: yonezawa@scphys.kyoto-u.ac.jp

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1 Magnetic susceptibility of CsV₃Sb₅

2 Figure S1a shows the temperature dependence of the magnetic susceptibility $\chi = M/H$ measured
3 under in-plane field of 7 T for both zero-field-cooling (ZFC) and field-cooling (FC) conditions. It
4 shows paramagnetic behaviour at high temperature. The magnetic susceptibility χ shows a sharp
5 drop at ~ 94 K, corresponding to the charge density wave (CDW) transition. At T_{CDW} , gaps open
6 on several parts of the Fermi surface and hence the density of states (DOS) at the Fermi level
7 drops, leading to the decrease of the Pauli susceptibility. The ZFC and FC data almost overlap
8 with each other. Figure S1b is the magnetic susceptibility measured at 5 Oe below 5 K. A sharp
9 superconducting transition is observed at $T_{\text{c}}^{\text{onset}} \sim 2.9$ K. These data are consistent with previous
10 studies^{1,2} and indicative of high quality of the sample.

11 Background subtraction for CsV₃Sb₅ sample #2

12 In our experiment, the Faraday effect of lens and fiber becomes significant under magnetic field
13 and contribute to the measured signal. In order to quantify the polar Kerr effect from the CDW
14 transition in CsV₃Sb₅, we used a Nb metal sheet, which is non-magnetic above $T_{\text{c}} \sim 9.2$ K, to
15 measure the background contribution. This measurement was done under experimental conditions
16 similar to the measurements of sample #2. Figure S2a shows the measured θ_{K} for Nb at 0.3 T.
17 We fit the data with a polynomial function to capture the temperature dependence and the resulted
18 curve is denoted as $\theta_{\text{K}}^{\text{Nb}}$. The background signal at a different magnetic field H is defined as $\theta_{\text{K}}^{\text{bg}} =$
19 $\theta_{\text{K}}^{\text{Nb}} \times \frac{\mu_0 H}{0.3} \times C$, where C is a constant determined by matching $\theta_{\text{K}}^{\text{bg}}$ with the data of CsV₃Sb₅ above

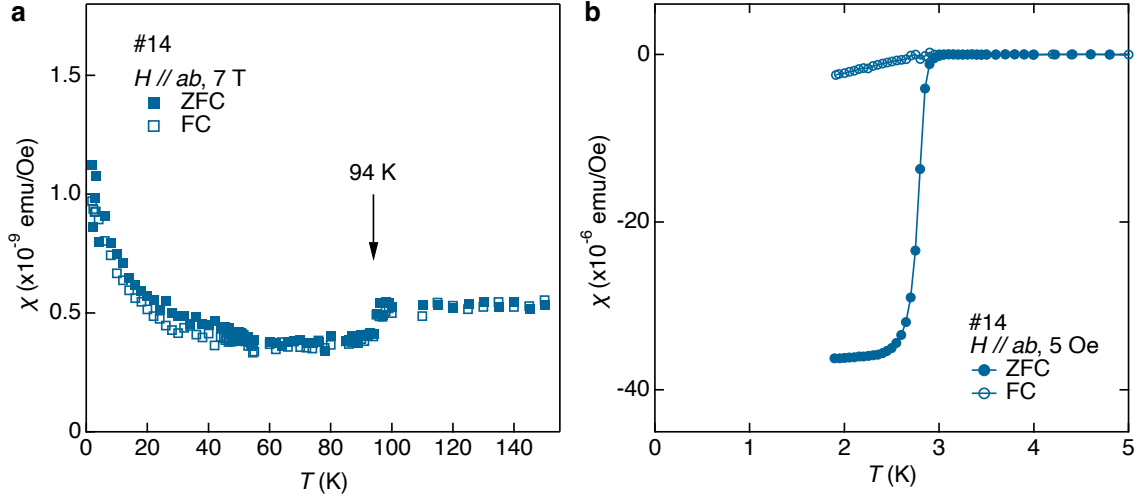


Figure S1: Temperature dependence of the magnetic susceptibility of CsV_3Sb_5 of sample #14. a.

Temperature dependence of the magnetic susceptibility χ measured at 7 T. The magnetic field is in the ab plane. The closed markers represent data collected at warming after zero-field-cooling (ZFC), while the open markers represent data measured during field-cooling (FC). The arrow indicates $T_{\text{CDW}} \sim 94$ K.

b. Temperature dependence of χ measured at 5 Oe parallel to the ab plane below 5 K, showing a sharp superconducting transition. The solid markers represent data collected at warming after zero-field-cooling (ZFC). The open markers represent data measured during field-cooling (FC).

T_{CDW} . We found that the constant C deviates from 1 typically by about 20%. This may be due
to the slight changes in the modulation frequencies and the positions of the lens and fiber in the
magnetic field. Figures S2b and c show θ_K of CsV_3Sb_5 measured at 0.3 T and 3 T together with
the background signal θ_K^{bg} . By subtracting θ_K^{bg} from θ_K , we obtain the background-subtracted polar
Kerr angle $\theta_K^{\text{sample}} = \theta_K - \theta_K^{\text{bg}}$, as shown in Fig. S2d and Fig. 2a in the main text. Note that in
Fig. S2c, the measured θ_K and θ_K^{bg} start to deviate below ~ 100 K and make θ_K^{sample} at 3 T above
 T_{CDW} increase. This is probably because the θ_K of Nb was measured at 0.3 T and may have slightly
different temperature dependence from θ_K at 3 T. Nevertheless, the small upturn in the θ_K^{sample} at 3 T
does not affect our observation that the polar Kerr angle from the sample changes at T_{CDW} and this
change can be switched by magnetic field.

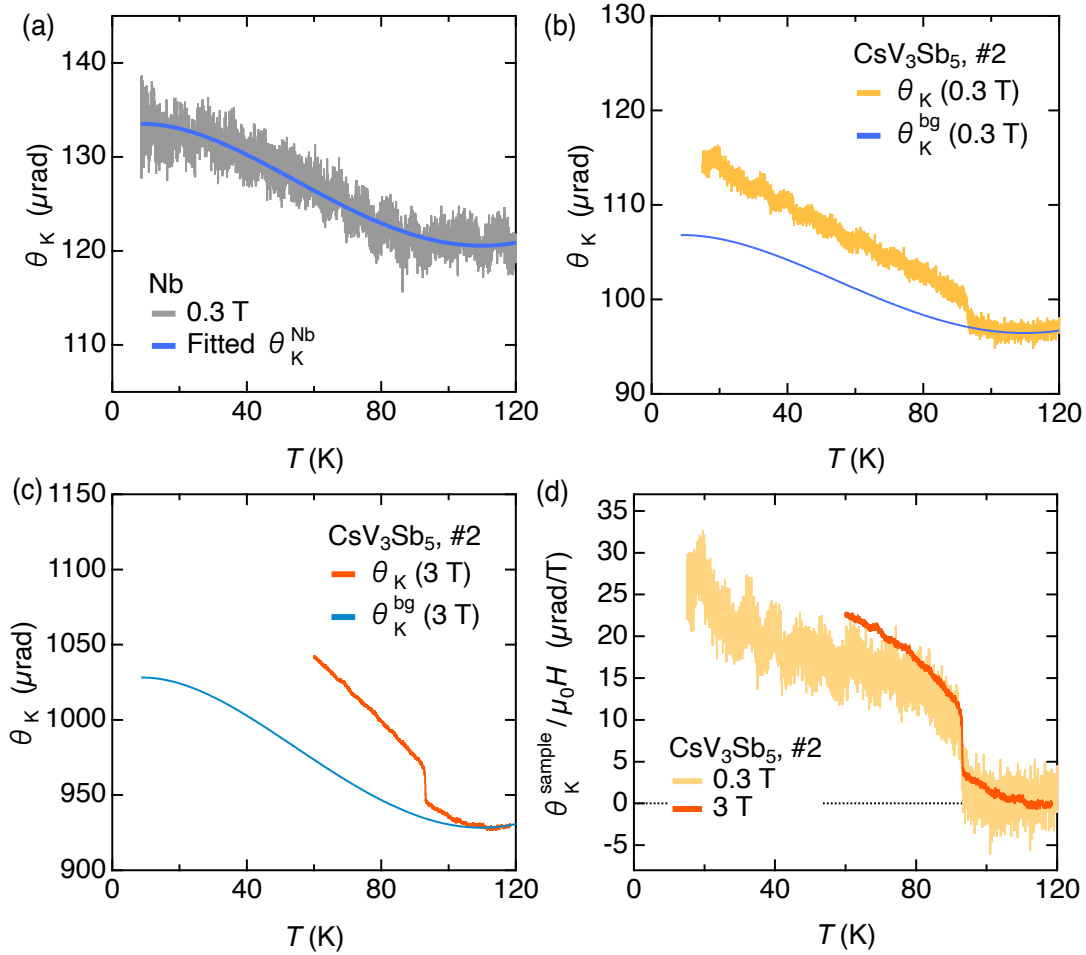


Figure S2: Examples of background subtraction to produce the data in Fig 2a of the main text, for sample #2. **a.** Temperature dependence of the polar Kerr angle θ_K of a Nb sheet measured at 0.3 T. The data is fitted by a polynomial function, as shown by the blue curve, θ_K^{Nb} . **b.** Temperature dependence of θ_K of CsV_3Sb_5 measured at 0.3 T. The background signal is θ_K^{bg} (0.3 T) = $\theta_K^{\text{Nb}} \times 0.8$. **c.** Temperature dependence of θ_K of CsV_3Sb_5 measured at 3 T. The background signal is θ_K^{bg} (3 T) = $\theta_K^{\text{Nb}} \times 10 \times 0.77$. **d.** Temperature dependence of the polar Kerr angle after background subtraction ($\theta_K - \theta_K^{\text{bg}}$) divided by magnetic field, the same as data shown in Fig. 2a of the main text.

Polar Kerr angle, second-harmonic signal and their temperature derivatives

In this section, we present the raw data used to extract the transition temperature T_{CDW} and polar Kerr angle jump $\Delta\theta_K$, presented in the inset of Fig. 1b and Fig. 2c of the main text. Among the three measured samples, the data are consistent.

Figure S3a shows the temperature derivative of the second-harmonic signal $V_{2\omega}$ (proportional to the sample reflectivity) near the CDW transition of #14. The data are collected from 1 T to 10 T. The sharp peaks around 93 K indicate the CDW transition. T_{CDW} is determined by the temperature where $|dV_{2\omega}/dT|$ peaks.

Figure S3b shows θ_K measured under negative magnetic field. At T_{CDW} , the polar Kerr angle shows a drop, which is symmetric to the data shown in Fig. 2b measured under positive field.

In the temperature dependence of θ_K , there is a tiny upturn (downturn) under positive (negative) magnetic field at ~ 95 K (Fig. 2b of the main text), implying an onset behaviour. We calculated the temperature derivative of the polar Kerr angle $|d\theta_K/dT|$, as shown in Fig. S3c, in order to investigate this behaviour more in detail. The sharp peaks indicate the jumps in θ_K at the CDW transition. Above 5 T, $|d\theta_K/dT|$ shows a broad hump above T_{CDW} . This feature may be attributed to multiple domains within the light spot area, which is not surprising for a CDW system exhibiting a first-order phase transition. Moreover, the hump becomes larger at higher magnetic field. If the hump above T_{CDW} comes from another domain which contribute only a tiny portion to the total signal, when the magnetic field is higher, θ_K is enhanced almost linearly and the hump

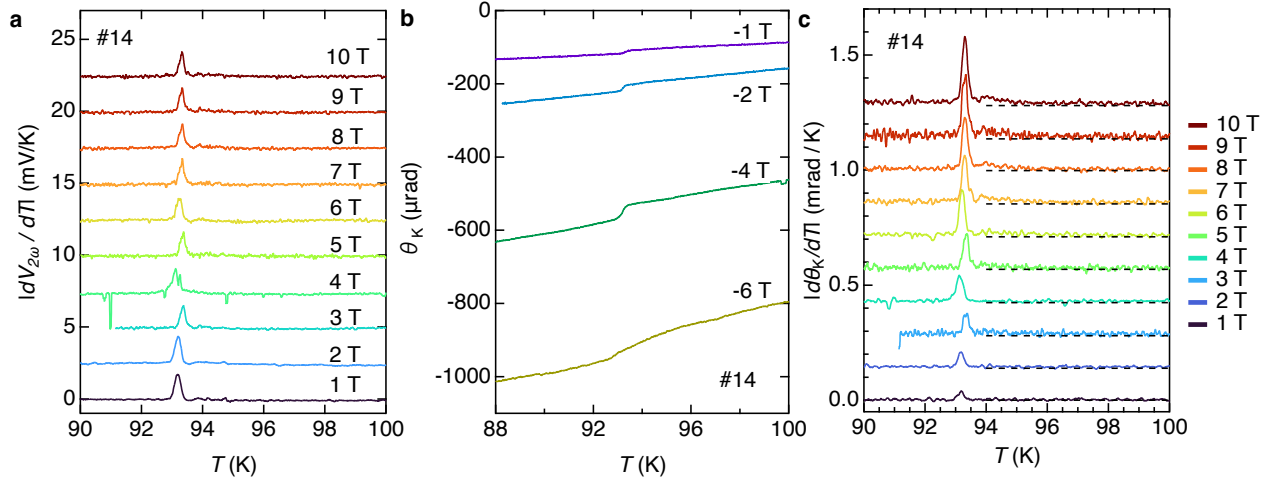


Figure S3: Temperature derivative of $V_{2\omega}$, θ_K , and its temperature derivative of sample #14. a.

Temperature dependence of the temperature derivative of $V_{2\omega}$ under magnetic field. The data are vertically offset for clarity. **b.** Temperature dependence of θ_K measured under negative magnetic field. **c.** Temperature dependence of $|d\theta_K/dT|$ under magnetic field. The data are offset for clarity. The dashed lines indicate the constant $|d\theta_K/dT|$ above T_{CDW} .

part becomes more visible.

Figure S4 shows the data of sample #10. The temperature derivative of the second-harmonic signal $|dV_{2\omega}/dT|$ shows a peak around 93 K, as shown in Fig. S4a. The data are used to obtain T_{CDW} up to 10 T (Fig. 1b). Figure S4b shows the temperature dependence of θ_K . For opposite magnetic fields, both the θ_K and the jump at T_{CDW} are flipped. The tiny kinks around 90 K in the ± 5 T data are due to drastic temperature change. Figure S5 shows the similar data measured on sample #2. The $|dV_{2\omega}/dT|$ shows peaks at T_{CDW} and the θ_K can be flipped by opposite magnetic fields.

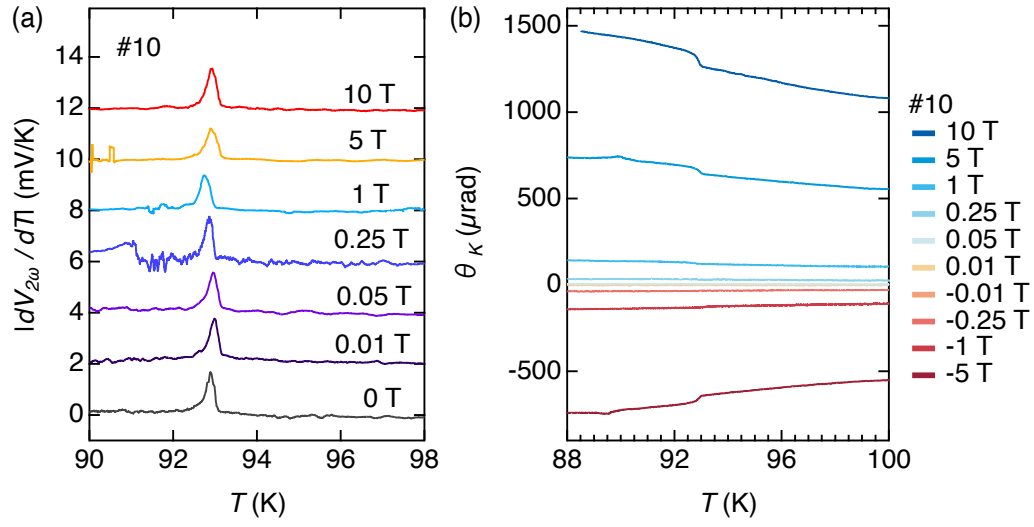


Figure S4: Temperature derivative of $V_{2\omega}$ and θ_K of sample #10. **a.** Temperature dependence of the temperature derivative of $V_{2\omega}$ under magnetic field. The data are vertically offset for clarity. **b.** Temperature dependence of θ_K measured under magnetic field.

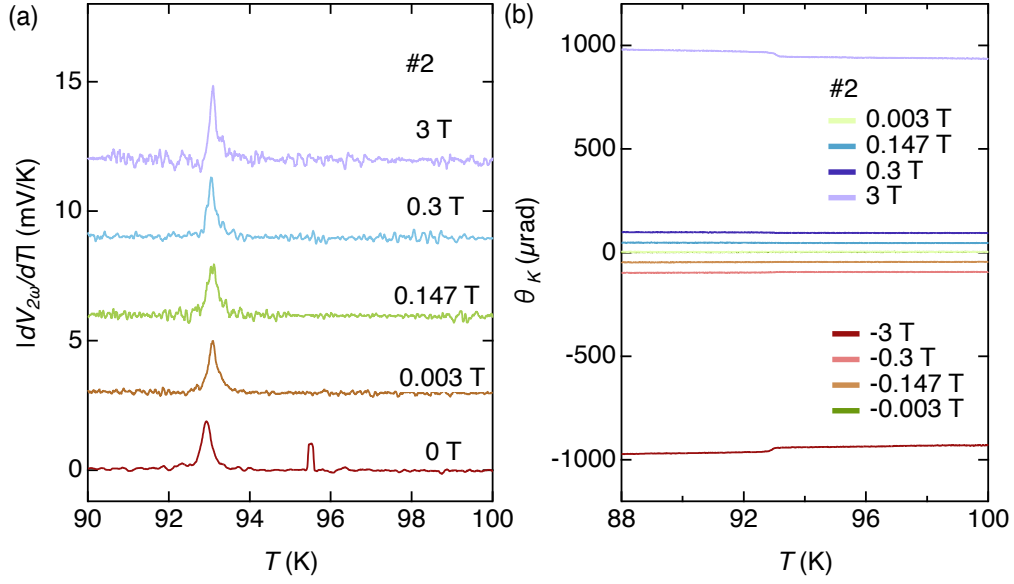


Figure S5: Temperature derivative of $V_{2\omega}$ and θ_K of sample #2. **a.** Temperature dependence of the temperature derivative of $V_{2\omega}$ under magnetic field. The data are vertically offset for clarity. **b.** Temperature dependence of θ_K measured under magnetic field.

Field-training measurements at different magnetic fields

Here, we present θ_K after different field-training, from which we extract the data in Fig. 4c of the main text. Figure S6 compares the temperature dependence of θ_K measured at zero-field-warming (ZFW) after field-cooling from above T_{CDW} with incident light power $\sim 200 \mu W$ and $\sim 100 \mu W$. All the data reproduce the field-training effect, showing a positive (negative) trend for positive (negative) training field.

For $200 \mu W$ measurements (Fig. S6a), the ± 2 T and 6 T data points are obtained by averaging the signal for 1 hour, while the ± 10 T data are obtained by 2-hour averaging. For $100 \mu W$ measurements (Fig. S6b), the data points are obtained by averaging the signal for 2 hours. We note that the fluctuation of $100 \mu W$ data is smaller than that of $200 \mu W$. This result contradicts the naive expectations and may be attributed to the domain effect and the heating effect. The light of smaller power effectively has smaller spot size. When the size of the light spot is comparable to the domain size, the spot of smaller power covers less domains and may reduce fluctuation. In addition, with smaller light power, the heating on sample surface is lower and may reduce thermal noise.

In Fig. S7, we present the temperature dependence of $V_{2\omega}$ (proportional to reflectivity) near T_{CDW} measured at 4 T with different light power. The data are normalized by $V_{2\omega}$ at 95 K. With decreasing power from $200 \mu W$ to $19 \mu W$, the CDW transition temperature T_{CDW} increases by around 1 K. Moreover, with smaller power, the transition tends to show two steps with a sharp jump at lower temperature and a broader jump at higher temperature. This feature is consistent with

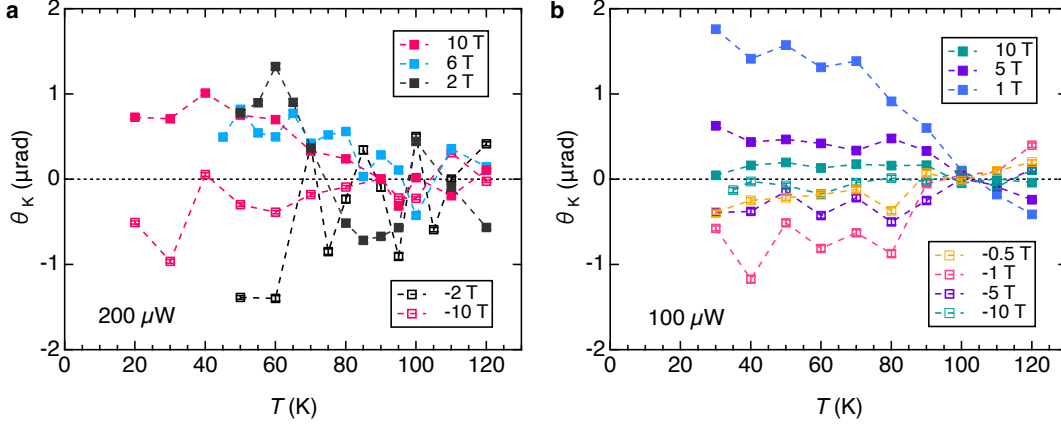


Figure S6: Temperature dependence of θ_K measured at zero-field-warming after field-training. **a.** θ_K measured with light power $\sim 200 \mu\text{W}$. **b.** θ_K measured with light power $\sim 100 \mu\text{W}$. A set of ± 1 T field training data shown in the main text is also plotted. The data are collected on sample #14.

the domain effect and the change of effective size of the light spot. For a smaller power (smaller effective spot size), a fewer number of domains are measured and the contribution from minor domains (with higher T_{CDW}) becomes more visible. Comparing the sharp jump at the transition, the difference of T_{CDW} between $200 \mu\text{W}$ and $100 \mu\text{W}$ is ~ 0.2 K. Thus, the heating effect should not be severe in our measurements.

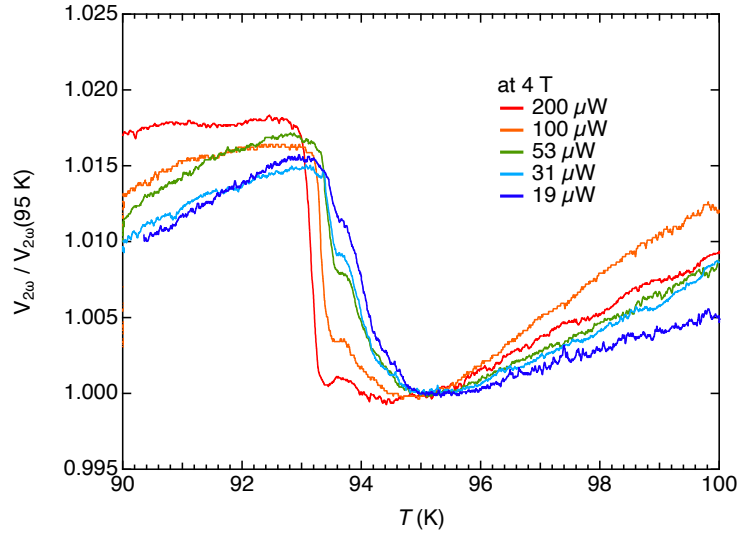


Figure S7: Temperature dependence of $V_{2\omega}$ near T_{CDW} with different light power. The data are collected on sample #14 at 4 T. $V_{2\omega}$ is normalized to $V_{2\omega}(95 \text{ K})$ in order to compare the transition temperature.

82 Reference

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