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Reply to "Comment on 'Charge-parity symmetry observed through Friedel oscillations in chiral charge-density waves' "

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We are responding to the Comment by J. Wezel on our paper. This study was developed from our previous work [Ishioka *et al.*, Phys. Rev. Lett. **105**, 176401 (2010)]. In the PRL paper, $H_{\rm CDW}$ was defined as a new parameter for expressing CDW chirality for the first time. In his Comment, he claims that $H_{\rm CDW}$ is ill defined. He also claims that the initial phase ϕ of the CDW wave function is a more appropriate parameter for expressing chiral CDW, despite our early introduction of ϕ to explain the experimental data described in the PRL paper. However, we conclude that $H_{\rm CDW}$ can distinguish the CDW chirality by its sign. Moreover, by considering different $H_{\rm CDW}$ signs, we had succeeded in demonstrating the difference of the spatial distributions of CDWs as shown in Fig. 4 of the PRB paper [Phys. Rev. B **84**, 245125 (2011)]. In our Reply, we discuss the validity of $H_{\rm CDW}$. We show that his argument regarding the identification of the CDW with the opposite sign of $\boldsymbol{q}$ is wrong, since the logic is inapplicable to a wave function with a nonzero ϕ. We also discuss the applicability of $H_{\rm CDW}$ to two- or three-dimensional CDWs in transition metal dichalcogenides.

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I. INTRODUCTION

As we reported in our PRL study,[1] in 1T-$TiSe_2$ we discovered chiral charge density waves (CDWs) for the first time by observing two-fold optical reflection and by detecting different CDW intensities for the q vectors of three CDWs. To explain the experimental results we introduced nonzero relative phases between three CDW wave functions. Right- and left-handed charge distributions in real space can be reproduced by choosing relevant phase differences. Moreover, to give the CDW chirality more generality, we focused on three-dimensional CDW q vectors and defined the CDW chirality as

$$H_{\rm CDW} \equiv \boldsymbol{q_1} \cdot (\boldsymbol{q_2} \times \boldsymbol{q_3}), \tag{1}$$

where $\boldsymbol{q_1}$, $\boldsymbol{q_2}$, and $\boldsymbol{q_3}$ are CDWs $\boldsymbol{q}$ vectors connecting Fermi pockets from Γ to L in the first Brillouin zone of 1T-$TiSe_2$. When $\boldsymbol{q_1}$, $\boldsymbol{q_2}$, and $\boldsymbol{q_3}$ do not exist in one plane, $H_{\rm CDW}$ has a nonzero value in the same way that cholesteric liquid crystals have a nonzero value of $\int \boldsymbol{n} \cdot (\nabla \times \boldsymbol{n}) dV$, where $\boldsymbol{n}$ is a director.[2] In spin systems, a similar form of scalar triple product of spins has been proposed as a chiral order parameter.[3] Since the mathematical form is the same, the introduction of $H_{\rm CDW}$ as a new measure is quite natural. As a measure for expressing the property of the entire space, it is highly intuitive to use a pseudoscalar.

In his Comment,[4] Wezel abandoned the $H_{\rm CDW}$ for the first time from the point of view that each q vector has invariance when translated by reciprocal lattice vectors so that the sign of the $H_{\rm CDW}$ changes arbitrarily. He insists that $H_{\rm CDW}$ has no physical meaning. However, we do not agree with this claim because this argument is not made for the identical initial phases of the three CDW wave functions.

II. FORBIDDEN OPERATION TO SPACE WITH CHIRALITY

In the first place, no arbitrary axis inversion can preserve an entire system that does not have mirror inversion symmetry. Let us consider a die representing systems without inversion symmetry. As shown in Fig. 1(a), we define each axis and assume a die. When we look at the 1, 2, and 3 faces of the die, they are located counter clockwise [Fig. 1(a)]. If the x axis is inverted, the arrangement of the 1, 2, and 3 faces becomes clockwise. The dice before and after axis inversion are not identical, that is the whole system changes in terms of chirality. The system with x- and y-axis inversion is the same as the original die [Fig. 1(c)]. As shown in Fig. 1(d), whole axis inversion changes the die's chirality. Therefore, as the property of the three-dimensional spaces, arbitrary axis inversion, namely mirror operation, inverts the entire space. In the same manner, when the pseudoscalar is defined, its sign should be changed by any mirror operation.

In general, a scalar triple product depends on the directions of triple q vectors. When there are three vectors, mirror inversion must make at least one vector inversion. On the other hand, in the PRB argument,[5] mirror inversion operation corresponds to the inversion of all three q vectors, when the mirror plane is perpendicular to the $\boldsymbol{c^*}$ axis. Because three vectors are inverted by mirror inversion, the scalar triple product of those vectors must change. When each vector characterizes the lattice structure, the whole lattice structure should be inverted in accordance with the vector inversion. Therefore, $H_{\rm CDW}$ reflects the chirality of the CDW structure. Indeed, the $H_{\rm CDW}$ value also changes from $-\frac{6\pi}{a^2c\sqrt{3}}$ to $\frac{6\pi}{a^2c\sqrt{3}}$. The $H_{\rm CDW}$ change corresponds to the handedness of the states.

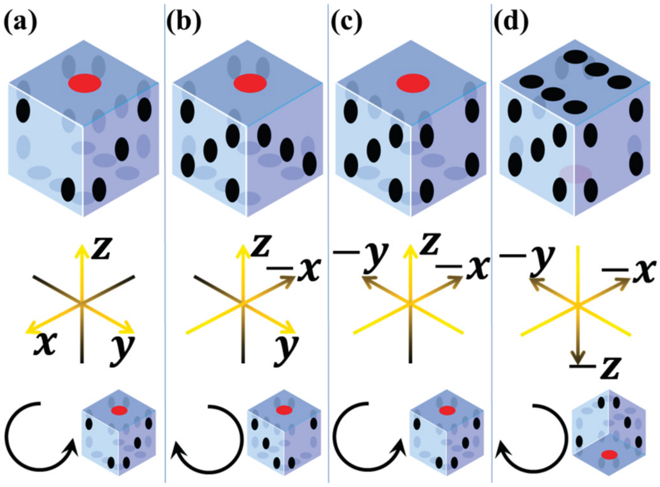

FIG. 1. (Color online) Schematic images of dice with (a) original structure, (b) x axis inverted, (c) x and y axis inverted and (d) x, y, and z axis inverted. The 1, 2, and 3 faces are shown at the bottom of each image.

III. CONCEPTUAL INCLUSION OF THE INITIAL PHASE DIFFERENCES BY H_{CDW}

The concept of the H_{CDW} includes the relative phase of the wave functions. Wezel's main claim is based on the invariance under the translation of each q vector. However, we think that the invariance of q is not demonstrated properly because he ignores the phase differences of each CDW although he insists on ϕ as an alternative. Within the very specific condition of $\phi = 0$ or π, the wave function with q is equal to that with $-q$. On the other hand, phase differences with more general values have been introduced to explain the experimental results as discussed in Ref. 1. The wave functions of CDW are expressed as

$$\rho = A\cos(qr + \phi) \tag{2}$$

and

$$\rho' = A\cos(-qr + \phi), \tag{3}$$

where ρ' is different from ρ with respect to the position of the charge density peaks. Therefore, a total configuration constructed with three CDW components of $-q_i$ should differ from that with q_i ($i = 1,2,3$). Since the formalization of the wave function[4,6] given by his results in a cosine type function, the above discussion can be applied to his discussion.

Returning to Wezel's claim, our discussion of the PRB paper is based on the fact mentioned above. In the numerical calculations shown in Fig. 4 in PRB[5] we first assume a set of three CDW waves and locally induced waves. Local chiral inversion was induced by the $\boldsymbol{q}$ inversion of only local components, and the two different states before and after q inversion were compared. If the invariance of $\boldsymbol{q}$ under arbitrary $\boldsymbol{a}^*$ remains with a nonzero ϕ as he insists, the spatial charge distribution of the two states must be the same. However, in three-dimensional spaces, there is no condition satisfying this. On the other hand, our calculation succeeded in showing the local chiral inversion by $\boldsymbol{q}$ inversion as the local modulation of the spatial distribution of the charge density. Therefore, in our research H_{CDW} clearly distinguishes two states with different chiralities.

He focuses solely on the phase difference between three CDWs. He also insists that the sign of ϕ corresponds to the chirality of the ground states. We agree with the correspondence, because the change in the sign of ϕ corresponds to the change in the sign of the q vector, that is,

$$\begin{aligned}\rho'' &= A\cos[qr + (-\phi)] = A\cos[-(qr + (-\phi))] \\ &= A\cos[-qr + \phi] = \rho'.\end{aligned} \tag{4}$$

So the chiral inversion could be expressed both by the phase difference and the q vectors. It is also possible to simulate the same calculation in Ref. 5 by changing signs of ϕ_i. To avoid any reader's confusion about our previous study,[1] we did not change the phases when all the $\boldsymbol{q}$ values were inverted in PRB.[5] Although ϕ_i is nonzero value, ϕ_i cannot distinguish whether CDW is *polar*[7] or *chiral*. A nonzero H_{CDW} clearly distinguishes tilted q vectors from in-plane q vectors. Therefore, we think the concept of H_{CDW} includes the sign of the phase ϕ so that H_{CDW} is a more sophisticated concept than ϕ for expressing chiral states.

IV. DIFFERENT APPROACHES TOWARDS THE SAME PHENOMENA

Regarding the paper on which Wezel's work was based,[6] we recognize it for its meaningful suggestion of the chiral state of CDWs based on a theoretical calculation of GL free energy.[6] This work strengthens our belief in the existence of chiral CDWs because it provides multiple explanations. However, to avoid confusing the readers, he should clarify the postulate of the theoretical analysis, which differs from that in our paper in PRL.[1] Based on experimental fact, we first fixed the phase difference between CDWs and then constructed H_{CDW} with q vectors. In this sense, the phase difference is a postulate for expressing chirality. On the other hand, he constructs GL free energy with ϕ as a variable and then calculates ϕ to minimize the total energy.[6] The calculation finally provides the nonzero phase differences of each CDW. Although different approaches were used, consistent results have been obtained in terms of chirality.

V. CONCLUSION AND REMARKS

In conclusion, we think that H_{CDW} is closely linked with the sign of the phase differences. It is quite difficult to directly detect phase differences. In contrast, H_{CDW} is highly intuitive to predict whether CDW have chirality by knowing the CDW q vectors. It will be used as a discriminant for chiral CDWs by testing whether H_{CDW} can be $(-,0,+)$. $-$, 0, and $+$ corresponds to left-handed chiral CDWs, nonchiral CDWs, and right-handed chiral CDWs, respectively.

Researchers have found magnetochiral anisotropy in the electric conductivity of carbon nanotubes and physically twisted metal wires.[8,9] This anisotropy depends on the structural chirality of each system. We expect a discussion to begin on a similar effect on chiral CDWs. When several systems show chiral CDWs and chiral properties, we believe H_{CDW} is more appropriate than just ϕ for discussing each effect universally because H_{CDW} directly reflects the structural characteristics including the size of the CDW superlattice.

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