Comment on "Low-temperature lattice excitation of icosahedral Al-Mn-Pd quasicrystals"

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By generalizing the wave propagation theory in crystalline solids to icosahedral quasicrystals (IQC's), Li and Liu [Phys. Rev. B **63**, 064203 (2001)] introduced the contribution of phasons to the lattice vibration of quasicrystals to interpret the distinctly large heat capacities of Al-Mn-Pd IQC at low temperature [Phys. Rev. B **57**, 10 504 (1998)]. However, we find that Li and Liu adopted several different coordinate systems for IQC's in their paper without any appropriate coordinate transformation. When the correct coordinate transformation is exploited, the calculated results disagree totally with the experimental ones.

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In order to explain the observed large deviation of the specific heat from Debye's law in $Al_{68.2}Mn_9Pd_{22.8}$ icosahedral quasicrystal (IQC) at low temperature,[1] Li and Liu[2] generalized the wave propagation theory in crystalline solids as continuous elastic medium to IQC's, by which they introduced the contribution of phasons to the low-temperature lattice vibration of IQC's. Based on that, they derived the generalized density of vibration states (DOVS) and specific heat expressions for Al-Mn-Pd IQC's. Then they obtained the numerical calculated values, which are in good agreement with the experimental data measured by Wälti *et al.*[1] Consequently, they concluded that it was the contribution of phasons that accounted for the excess specific heat values for Al-Mn-Pd IQC at low temperature. However, we would point out that they directly employed the elastic constants for IQC's, both the form and quantities, from several different coordinate systems without coordinate transformation, which made their calculation totally wrong and their conclusion unconvincing.

As we know, to apply the generalized elasticity theory to the case of IQC's, one should note that the elastic constants depend on the choice of the coordinate system, on the concrete matrix describing the projection of the six-dimensional (6D) hyperspace basis vectors into the 3D physical space, and even on the personal notation of the elastic constants.[3–5] Unfortunately, lacking a standard choice like in crystals, the researchers chose a particular coordinate system for IQC's somewhat arbitrarily for their own convenience.[5–22] The coordinate systems in the literature can be roughly classified into two groups. One type of coordinate system can be chosen with the z axis (i.e., $\mathbf{E}_3^{\parallel}$ in Fig. 1) pointing towards a vertex of an icosahedron (i.e., along the fivefold axis).[6–9] Another type of coordinate system has the z axis normal to an edge of an icosahedron (i.e., along the twofold axis).[5,10–22] In each case, there also exist different concrete projection matrices and notation of elastic constants, which makes it more complex and more confusing to apply the elasticity theory for IQC's. We will discuss these in detail in another paper.[4]

In the paper by Li and Liu,[2] the authors used the linear elasticity theory for IQC's and elastic constant form, given by Ding *et al.*,[6,7] to derive expressions for wave propagation in IQC's. However, they adopted the projection matrix in another coordinate system [Eq. (22) in Ref. 2], provided by Elser,[10] to get the corresponding direction cosines (l,m,n) in 3D physical space from the wave propagation direction vectors in 6D hyperspace. As mentioned above, the coordinate system, used to get the elastic constants form by Ding *et al.*,[6,7] belongs to the first group. However, the coordinate system corresponding to the projection matrix, provided by Elser,[10] is in the second group. So the direction cosines (l,m,n) derived by Li and Liu are inappropriate for their expressions obtained on the basis of the elastic constant form of IQC's provided by Ding *et al.* We show the coordinate system in the physical space adopted by Ding *et al.*[6,7] as Fig. 1. Correspondently, Eq. (22) for the projection matrix in Ref. 2 should be replaced by the following one, which is suitable for the system adopted by Ding *et al.*[6,7]

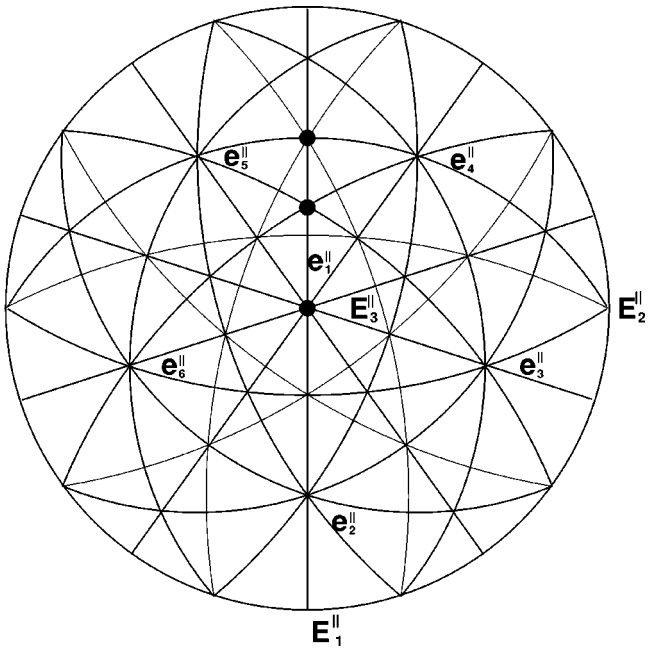

FIG. 1. The stereographic projection showing the coordinate system in the 3D physical space adopted by Ding *et al.* (Refs. 6 and 7), where $\mathbf{E}_1^{\parallel}$, $\mathbf{E}_2^{\parallel}$, $\mathbf{E}_3^{\parallel}$ are the basis vectors in physical space and $\mathbf{e}_i^{\parallel}$ $(i=1,2,\ldots,6)$ are the projection of the basis vectors from the 6D hyperspace to the physical space. The three solid circles denote the currently studied wave propagation directions, i.e., fivefold, twofold, and threefold axis directions.

TABLE I. The velocities of the acoustic phonons and phasons of the icosahedral $Al_{68.2}Mn_9Pd_{22.8}$ quasicrystal along fivefold, twofold, and threefold axis directions respectively. The original values of elastic constants used here are the same as those adopted by Li and Liu (Ref. 2); however, we applied indispensable coordinate transformation, rather than used them directly, before the calculation (see text for details).

Axis	Direction	v_1 (m/s)	v_2 (m/s)	v_3 (m/s)	v_4 (m/s)	v_5 (m/s)	v_6 (m/s)
*A*5	(1,0,0,0,0,0)	6340.5	3570.0	3570.0	3067.8	3067.8	5386.4
*A*2	(1,1,0,0,0,0)	6340.1	3570.0	3570.0	1830.8	4383.6	5027.5
*A*3	(1,1,1,1,1,1)	6340.0	3564.1	3575.3	1695.8	4733.3	4750.6

$$\tilde{Q}_{||}^{-1}=\frac{1}{\sqrt{2}}\begin{pmatrix} 0 & 0 & 1 \\ S & 0 & C \\ S\cos\theta & S\sin\theta & C \\ S\cos 2\theta & S\sin 2\theta & C \\ S\cos 3\theta & S\sin 3\theta & C \\ S\cos 4\theta & S\sin 4\theta & C \end{pmatrix}, \qquad (1)$$

where $\theta=2\pi/5$, $S=\sin 63.4°=2/\sqrt{5}$, and $C=\cos 63.4°=1/\sqrt{5}$. Then the correct direction cosine (l,m,n) values can be obtained. For example, the cosines for the fivefold direction can be obtained as $l=0,m=0$, and $n=1$, rather than $l=\tau/\sqrt{1+\tau^2}$, $m=0$, and $n=1/\sqrt{1+\tau^2}$, projected from the 6D lattice vector (1,0,0,0,0,0) with this correct projection matrix. Here, $\tau=(1+\sqrt{5})/2$ is the golden mean. The solid circles in Fig. 1 show the three currently studied directions, which correspond to the 6D lattice vectors (1,0,0,0,0,0), $(1,\bar{1},0,0,0,0)$ and $(1,1,\bar{1},1,1,\bar{1})$, respectively, in the stereographic projection pattern of IQC's in physical space.

Furthermore, Li and Liu[2] took the following values of elastic constants from other papers[19–22] for their numerical calculations: $\lambda=0.75$, $\mu=0.65$, $K_1=0.81$, $K_2=-0.50$, and $R=0.0066$ (10^{12} dyn/cm^2). However, Li and Liu substituted them in their expressions directly, without appropriate coordinate transformation. No problem exists for the Lamé constants λ and μ. Unfortunately, it is not the case for the others. We know that Refs. 20–22, which Li and Liu referred to for choosing the values of K_1, K_2, and R, adopted the same coordinate system as Jarić *et al.*[13] and Widom,[14] and used the concrete notation of elastic constants defined by Widom.[14] The relationships between the elastic constants in the coordinate system used by Ding *et al.*[6,7] (denoted by the superscript D) and those by Widom[14] (denoted by the superscript W) are[4] $K_1^{\mathrm{D}}=K_1^{\mathrm{W}}-K_2^{\mathrm{W}}/3$, $K_2^{\mathrm{D}}=-K_2^{\mathrm{W}}$, and $R^{\mathrm{D}}=K_3^{\mathrm{W}}$. Using these correct transformations, the above elastic constant values should be transformed to $K_1^{\mathrm{D}}=0.98$, $K_2^{\mathrm{D}}=0.50$, and $R^{\mathrm{D}}=0.0066$ (10^{12} dyn/cm^2) before substituting them into the expressions.

With the correct projection matrix and elastic constant values, we calculated the velocities of the acoustic phonons and phasons, the coefficients of the DOVS, and the specific heat C_{ph} of $Al_{68.2}Mn_9Pd_{22.8}$ IQC, by following the theory suggested by Li and Liu.[2] All the values are calculated with the aid of the symbolic manipulation program MAPLE (Ref. 23) and shown in Table I and Table II, respectively.

From Table I, we can see that the values of v_i ($i=1,2,3$) are nearly the same as the results given by Ref. 2 and are in good agreement with the results of the resonant ultrasound spectroscopy experiment[19] indeed, which is a natural result for the very small phonon-phason coupling. However, the other values in Table I show obvious anisotropy and are completely different from those in Ref. 2, which directly influence the values of a, b, β, and δ as shown in Table II. Moreover, Li and Liu[2] argued that the small anisotropy of the values in their Table I might be the reason that the IQC's have the small anisotropy of the temperature dependence of the magnetoresistivity observed in the experiment of Rodmar *et al.*[24] However, we would like to point out here that Rodmar *et al.*[25] have concluded that the temperature dependence of the magnetoresistance is isotropic and some small variation could be correlated to a gradient of the Mn concentration of below 0.2%/cm in the growth direction of the quasicrystal grain after careful examinations.

As shown in Table II, when the correct transformation matrix and elastic constant values are used, the values of

TABLE II. The coefficients of DOVS and C_{ph} of the icosahedral $Al_{68.2}Mn_9Pd_{22.8}$ quasicrystal from the experiment measurement (Ref. 1) and calculated values based on the theoretical model suggested in Ref. 2 after correct coordinate transformation, where the theoretical calculated values Calc. *A*5, *A*2, and *A*3 are along fivefold, twofold, and threefold axis directions, respectively.

	DOVS		C_{ph}			
	a (s^3/rad^3 mol)	b (s^5/rad^5 mol)	β (J/mol K^4)	δ (J/mol K^6)	ω_0 (rad/s)	Θ_{D} (K)
Expt. data	3.27×10^{-17}	2.37×10^{-43}	2.63×10^{-5}	9.21×10^{-8}	3.148×10^{13}	420
Calc. *A*5	2.92×10^{-17}	1.58×10^{-43}	2.35×10^{-5}	6.15×10^{-8}	3.396×10^{13}	436
Calc. *A*2	5.45×10^{-17}	12.79×10^{-43}	4.39×10^{-5}	49.75×10^{-8}	2.284×10^{13}	354
Calc. *A*3	6.42×10^{-17}	19.92×10^{-43}	5.17×10^{-5}	77.48×10^{-8}	2.095×10^{13}	335

a, b, β, and δ are completely different from those given by Li and Liu[2] and disagree totally with the experimental ones of Ref. 1. If we plot the calculated $C_{ph}/T^3=\beta+\delta T^2$ values, using the calculated β, δ in Table II, versus T^2 with the same scale of Fig. 4 in Ref. 1, in which the experimental values are provided, we can see the big discrepancy between the calculated and the experimental values directly. The values along twofold and threefold axis are much bigger than anticipated.

In conclusion, we find that there is a severe mistake in Ref. 2, in which the authors were unaware of the difference between different coordinate systems for IQC's and used both expressions and values for the elastic constants from different systems together, without appropriate transformation. When the correct projection matrix and appropriate transformed elastic constant values are adopted, the calculated coefficients of the DOVS and C_{ph} of the Al-Mn-Pd IQC's based on their theoretical model do not agree with the experimental results at all. This disagreement indicates that the theory suggested by Li and Liu[2] is inappropriate for describing the deviation of the specific heat of Al-Mn-Pd IQC's at low temperature from Debye's law in the present form. To improve this theory, we also suggest more consideration in choosing more appropriate values of the elastic constants for the calculation, besides the correct projection matrix and the indispensable coordinate transformation for elastic constants given in this paper.

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