Reply to “Comment on ‘Low-temperature lattice excitation of icosahedral Al-Mn-Pd quasicrystals’”

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In response to the comment by Wang and Qin, we have rewritten our equation (22) following the coordinate system of Ding et al. [J. Wuhan Univ. (Nature Science Edition) 3, 23 (1992)]. According to the thermodynamic stability condition and the experiment of Capitan et al., we have chosen the new parameters of the phason elastic constants. Based on this, we have recalculated the coefficients of the expressions on the vibrational density of states and the specific heat of the icosahedral Al-Mn-Pd quasicrystal. Our results are still in agreement with the experimental data measured by Wälti et al. [Phys. Rev. B 57, 10 504 (1998)]. It demonstrates that our method is useful for dealing with the low-temperature vibrational excitation of icosahedral quasicrystals.

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In order to reply the comment by Wang and Qin, we have examined the relationships between the coordinate system used by Widom’ and that used by Ding et al. In Widom’s coordinate system, the six basic vectors of the icosahedral quasicrystal are used by Widom1 and that used by Ding examined the relationships between the coordinate system et al.2,5 they can be described as

\[ \vec{e}_1 = \eta(\tau, 0, 1), \]
\[ \vec{e}_2 = \eta(\tau, 0, -1), \]
\[ \vec{e}_3 = \eta(1, \tau, 0), \]
\[ \vec{e}_4 = \eta(0, 1, \tau), \]
\[ \vec{e}_5 = \eta(0, -1, \tau), \]
\[ \vec{e}_6 = \eta(1, -\tau, 0), \]

where \( \eta = 1/\sqrt{2\tau + 4} \). For the coordinate system of Ding et al.,2,5 they can be described as

\[ \vec{e}_1 = G(0, 0, 1), \]
\[ \vec{e}_2 = G(\sin \beta, 0, \cos \beta), \]
\[ \vec{e}_3 = G(\sin \beta \cos \theta, \sin \beta \sin \theta, \cos \beta), \]
\[ \vec{e}_4 = G(\sin \beta \cos 2\theta, \sin \beta \sin 2\theta, \cos \beta), \]

where \( G = 1/\sqrt{2}, \theta = 2\pi/5, \sin \beta = \sin 63.4^\circ = 2/\sqrt{5}, \) and \( \cos \beta = \cos 63.4^\circ = 1/\sqrt{5} \). Comparing with the requirement of Eqs. (6)–(9) in Ref. 6, we find that the coordinate transformation and the relationships between the phason elastic constant of Ding et al. and Widom’s elastic constant given by Wang and Qin should be adopted. Consequently, Eq. (22) of the projection matrix in Ref. 6 should be rewritten as

\[ \vec{Q}_1^{-1} = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 0 & 1 \\ \sin \beta & 0 & \cos \beta \\ \sin \beta \cos \theta & \sin \beta \sin \theta & \cos \beta \end{pmatrix} \]

Then the correct direction cosine (l, m, n) values of the fivefold axis (1,0,0,0,0,0), twofold axis (1, -1, 0, 0, 0, 0), and threefold axis (1, 1, -1, 1, 1, -1) are, respectively,

\[ (0,0,1), \left( -\frac{1}{\sqrt{3}-\tau}, 0, \frac{1}{\sqrt{3}-\tau} \right). \]

TABLE I. Coefficient parameters and velocities of the acoustic phonons and phasons of icosahedral Al_{68.2}Mn_{9}Pd_{22.8} quasicrystal in the range [\eta]<0.35 Å⁻¹.

<table>
<thead>
<tr>
<th>Axis</th>
<th>Direction</th>
<th>( v_1 ) (m/s)</th>
<th>( \eta \times 10^{-17} ) (m²/s)</th>
<th>( v_2 ) (m/s)</th>
<th>( v_3 ) (m/s)</th>
<th>( v_4 ) (m/s)</th>
<th>( v_5 ) (m/s)</th>
<th>( v_6 ) (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>(1,0,0,0,0,0)</td>
<td>6340.2</td>
<td>-3.9</td>
<td>3570.0</td>
<td>3570.0</td>
<td>2425.0</td>
<td>5048.8</td>
<td>5048.8</td>
</tr>
<tr>
<td>2</td>
<td>(1,-1,0,0,0,0)</td>
<td>6348.3</td>
<td>-3.9</td>
<td>3574.8</td>
<td>3574.4</td>
<td>2550.6</td>
<td>4733.9</td>
<td>5337.3</td>
</tr>
<tr>
<td>3</td>
<td>(1,1,-1,1,1,-1)</td>
<td>6341.6</td>
<td>-3.9</td>
<td>3570.6</td>
<td>3570.7</td>
<td>2425.7</td>
<td>5063.5</td>
<td>5062.7</td>
</tr>
</tbody>
</table>

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TABLE II. Coefficients of DOVS and \( C_{\text{ph}} \) obtained from the theoretical calculation and the experiment measurement (Ref. 9) where the theoretical value 1 (thor. 1) (Ref. 9) stands for results only containing the contribution of acoustic phonons. The theoretical values 2–4 do for the results containing the contributions of acoustic phonons, acoustic phasons, and phonon-phason coupling to DOVS or lattice specific heat and they have been calculated by using the phase velocities along the five fold, two fold, and three fold directions respectively.

<table>
<thead>
<tr>
<th>DOVS</th>
<th>( a (s^2/\text{rad}^2 \text{mol}) )</th>
<th>( b (s^2/\text{rad}^2 \text{mol}) )</th>
<th>( C_{\text{ph}} )</th>
<th>( \beta (\text{J/mol K}^2) )</th>
<th>( \delta (\text{J/mol K}^6) )</th>
<th>( \Theta_D (\text{K}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Expt.</td>
<td>( 3.27 \times 10^{-17} )</td>
<td>( 2.37 \times 10^{-43} )</td>
<td>( 2.63 \times 10^{-5} )</td>
<td>( 9.2 \times 10^{-8} )</td>
<td>420</td>
<td></td>
</tr>
<tr>
<td>Theor.1</td>
<td>( 1.8 \times 10^{-17} )</td>
<td>( 0.65 \times 10^{-43} )</td>
<td>( 1.63 \times 10^{-5} )</td>
<td>( 2.5 \times 10^{-8} )</td>
<td>492</td>
<td></td>
</tr>
<tr>
<td>Theor.2</td>
<td>( 3.16 \times 10^{-17} )</td>
<td>( 2.77 \times 10^{-43} )</td>
<td>( 2.54 \times 10^{-5} )</td>
<td>( 10.8 \times 10^{-8} )</td>
<td>424</td>
<td></td>
</tr>
<tr>
<td>Theor.3</td>
<td>( 2.93 \times 10^{-17} )</td>
<td>( 2.18 \times 10^{-43} )</td>
<td>( 2.36 \times 10^{-5} )</td>
<td>( 8.49 \times 10^{-8} )</td>
<td>434</td>
<td></td>
</tr>
<tr>
<td>Theor.4</td>
<td>( 3.09 \times 10^{-17} )</td>
<td>( 2.62 \times 10^{-43} )</td>
<td>( 2.49 \times 10^{-5} )</td>
<td>( 10.2 \times 10^{-8} )</td>
<td>427</td>
<td></td>
</tr>
</tbody>
</table>

and

\[
\left( -\frac{2}{\sqrt{4+\tau^2}}, \frac{\tau^2}{\sqrt{4+\tau^2}} \right),
\]

where \( \tau = (1 + \sqrt{5})/2 \) is the “gold mean” of the Fibonacci sequence. Furthermore, the relationships between the elastic constants in the coordinate system used by Ding et al. (denoted by the superscript \( D \)) and those by Widom (denoted by the superscript \( W \)), as pointed out by Wang and Qin, are

\[
K_1^D = K_1^W - K_2^W/3, \quad K_2^D = -K_2^W, \quad K_3^D = K_3^W.
\]

However, the values of \( K_1^D = 0.98, \quad K_2^D = 0.5 \), chosen by Wang and Qin, are no longer suitable because they do not satisfy the thermodynamic stability condition

\[
K_1^D - 2K_2^D = 0.
\]

Therefore, the new values satisfying the thermodynamic stability condition must be retaken. On this point, Wang and Qin have the same opinion as us (private communication). According to the experiment of Capitan et al. and Widom’s theory, in the fivefold axis direction, we should take \( K_1^W = 0.97, \quad K_2^W = 0.5, \) then, \( K_3^D = 0.80, \quad K_1^D = -0.5, \) and \( R_D = 0.0066(10^{12} \text{ dyn/cm}^2) \). In the two- and threefold directions, we should take \( K_1^W = 0.97, \quad K_2^W = -0.5, \) and \( K_3^W = 0.0066 \) then, \( K_1^D = 1.14, \quad K_2^D = 0.5, \) and \( R_D = 0.0066(10^{12} \text{ dyn/cm}^2) \). For the phonon elastic constants, we can still choose \( \lambda = 0.75, \quad \mu = 0.65 \) \( (10^{12} \text{ dyn/cm}^2) \). Inserting these new values into Eqs. (14–(16) and then into Eq. (20) in Ref. 6, we obtain the velocities of the acoustic phonons and phasons and list them in Table I. In addition to substituting the calculated data of velocities into the coefficient equations (26), (28), (31), and (32) of the DOVS and the specific heat \( C_{\text{ph}} \) of Al\(_{68.2}\)Mn\(_{9}\)Pd\(_{22.8}\) icosahedral quasicrystal in Ref. 6, we work out all coefficients and list them in Table II.

The comparison with the data in Tables I and II of Ref. 6 shows that the new results in the present Tables I and II have only a small change and that those of them containing the contribution of phonons, phasons, and phonon-phason coupling still agree with the measured data by Wälti et al. Evidently, the theoretical values without the contribution of the phasons are obviously smaller than the experiment data, in which the coefficient of \( T^2 \) or \( T^3 \) term is almost half of the experiment data and that of \( T^4 \) or \( T^5 \) term is about one-fifth of the experiment data. Our present results demonstrate that the contribution of phasons cannot be neglected, and the higher the order of DOVS or specific heat term is, the stronger its affection is.

In Ref. 6 we had a try at explaining the puzzle that the observed values exceed the Debye prediction by the extensive Debye model. At present, our theory is still a phenomenological theory.

We would like to thank Dr. Wang and Dr. Qin for their helpful comments making the problems studied clearer.

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