

# Low-temperature lattice excitation of icosahedral Al-Mn-Pd quasicrystals

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(Received 11 July 2000; published 19 January 2001)

We suggest a theoretical method for dealing with the contribution of phasons to the lattice vibration of quasicrystals. Based on this, we derive the density of vibrational states (DOVS) and specific-heat expressions of the icosahedral Al-Mn-Pd quasicrystal, and obtain corresponding numerical solutions which are in agreement with the experimental data measured by Wälti *et al.* [Phys. Rev. B **57**, 10 504 (1998)]. This consistency shows that the contribution of the phasons to either the DOVS or specific heat cannot be neglected at low temperature. Our theory would be also helpful to study further the thermal conductivity of the icosahedral quasicrystal.

DOI: 10.1103/PhysRevB.63.064203

PACS number(s): 61.44.Br, 05.70.Ce, 62.20.Dc

## I. INTRODUCTION

Quasicrystals are a form of solids different from both crystals and glasses by possessing long-range noncrystallographical orientational order and a type of long-range translational order, quasiperiodicity.<sup>1</sup> The icosahedral quasicrystals studied intensively are quasiperiodic in all three dimensions. They display some very unusual physical properties: a very low electrical conductivity, a negative temperature coefficient of resistivity, a low electronic contribution to the specific heat, and so on.<sup>2-4</sup> Another experiment<sup>5</sup> exhibits the fact that icosahedral quasicrystals possess a small anisotropy of the temperature dependence of magnetoresistivity, which is different from the current theory proving that icosahedral quasicrystals have isotropic physical properties. Above all, recent experiments<sup>6-11</sup> on the heat capacities discovered that there exist excess heat capacities in quasicrystals at low temperature. Some of them are the stable icosahedral Al-Mn-Pd and Al-Cu-Fe quasicrystals which do not show a phason broadening of the Bragg peaks even in the nonannealed states.<sup>12,13</sup> Structure analysis indicates that it forms an ordered icosahedral state. In studying its lattice excitations, Jasjaunias *et al.*<sup>14</sup> and Wälti *et al.*<sup>15</sup> found that the cubic-in- $T$  term coefficient of the low-temperature lattice specific heat,  $C_{ph}(T)$ , is considerably higher than the Debye acoustic phonon contribution. Above 5 K, the ratio  $C_{ph}/T^3$  increases rapidly with increasing  $T$ . To explain the experimental results, Lasjaunias *et al.*<sup>14</sup> and Wälti *et al.*<sup>15</sup> considered that the deviations from the Debye model could be treated by putting in an additional term as  $\delta T^5$  which corresponds to an increase of the density of vibrational states (DOVS) more rapid than  $\omega^2$ , so  $g(\omega) = a\omega^2 + b\omega^4 + \dots$ , and then  $C_{ph} = \beta T^3 + \delta T^5$ . For further treatment, the above two groups had different ideas. Lasjaunias *et al.* suggested a new power law of specific heat as  $T^{3.6}$  (or  $T^{3.9}$ ) for AlCuFe<sub>12.5</sub> (or AlCuFe<sub>12</sub>) over one decade of temperature. In another way, Wälti *et al.* kept the  $T^5$  term, but by the use of experiment data fit the coefficients  $\beta$  and  $\delta$ . Their treatments, certainly, use phenomenological theory and have not revealed the physical ori-

gins of the deviation in specific heat yet. In the present article we suggest an alternative explanation, even though in the present stage it is also a phenomenological treatment. We note that in the works of the above two groups they considered only the contribution of acoustic phonons to the DOVS and  $C_{ph}$  of the icosahedral quasicrystal. As we know, besides the phonon, the phason is one of the main features which distinguish quasicrystals from crystals, so it must be of influence on the physical properties of quasicrystals such as the DOVS, specific heat, thermal conductivity, etc. About the phason, there are a few kinds of different points of view. According to the unit-cell picture,<sup>16</sup> the phason in quasicrystals is regarded as rearrangements of atoms from one lattice pattern to another, so it should be equivalent to the local defects with only short-range correlation. According to Lubensky *et al.*,<sup>17</sup> the phason modes represent the relative motion of the constituent density waves. Based on the fact that  $\nabla \cdot \mathbf{w}$  is not a scalar under the icosahedral group, they claimed that phasons are diffusive with very large diffusion times. According to Bak,<sup>18</sup> the phason describes particular structural disorders or structure fluctuations in quasicrystals, and it can be formulated based on a six-dimensional space description. For the icosahedral quasicrystals, the structure fluctuations can be characterized by three bulk translation modes and three relative phase-shift modes associated with internal atomic position rearrangements. These modes are, respectively, described in parallel and the perpendicular spaces. Spatially varying displacements in the parallel space are described by the phonon field  $\mathbf{u}$ , while that in the perpendicular space by the phason field  $\mathbf{w}$ . Spatially uniform displacements in both spaces leave the system free energy invariant. Since there are six continuous symmetries, there exist six hydrodynamic vibration modes. In this paper, we follow Bak's argument to describe the phonons and phasons in quasicrystals and assume that the phason fluctuations can produce long-wavelength elastic waves and the waves can propagate in real space. Along this line, Jeong and Steinhart<sup>19</sup> proved that in the unlocked phase, the phasons have a thermal excitation analogous to phonons and calcu-

lated the elastic constants corresponding the phason strains by using Monte Carlo simulations. In 1999, Naumis *et al.*<sup>20</sup> showed that a random phason produced in hyperspace leads to a coherent phason field in real space. Fan<sup>21</sup> calculated the phonon velocities in a plane field and phason velocities in an antiplane field of one-dimensional hexagonal quasicrystals. Then, based on our equivalent assumption of phonons and phasons, we will use the quasicrystal continuous model and linear elastic theory to calculate the phonon and phason phase velocities of the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal. By adding the phason velocities into the DOVS expression, we explain the DOVS and specific-heat experiment performed on the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal. Our results show that only acoustic phonons are not enough to describe the thermodynamical properties of quasicrystals. In other words, the contribution of the phason excitations to the DOVS or specific heat cannot be neglected at low temperature.

We organize our paper as follows: Section II presents linear elasticity theory, the elastic wave propagating model, and an approach calculated effective elasticity constant of the icosahedral quasicrystals. In Sec. III we derive these expressions of the phase velocities, DOVS, and lattice specific heat. Besides this, we numerically calculate the phase velocities and the coefficients of the DOVS and lattice specific heat in the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystals. In Sec. IV, we compare our theoretical results with the experimental data measured by Wälti *et al.*<sup>15</sup> Section V gives a brief summary.

## II. LINEAR ELASTICITY THEORY AND CONTINUOUS MODEL OF ICOSAHEDRAL QUASICRYSTALS

We now use the projection method to describe a phonon and a phason, and calculate their phase velocities by using linear elasticity theory. According to the projection method, an icosahedral quasicrystal can be obtained by projecting a six-dimensional periodic lattice onto a three-dimensional (3D) physical subspace.<sup>22–24</sup> Letting  $\vec{\xi}$  be a displacement vector in the six-dimensional space,  $\mathbf{u}$  and  $\mathbf{w}$  be two components of  $\vec{\xi}$  in the parallel subspace (i.e., 3D physical subspace  $V_{\parallel}$ ) and perpendicular subspace (i.e., 3D complementary subspace  $V_{\perp}$ ), respectively, then

$$\vec{\xi} = \mathbf{u} + \mathbf{w}, \quad (1)$$

where  $\mathbf{u}$  and  $\mathbf{w}$  are referred to as the phonon field and phason field, respectively. With  $u_1, u_2, u_3$  denoting the displacement components of the phonon field  $\mathbf{u}$  and  $w_1, w_2, w_3$  denoting those of the phason field  $\mathbf{w}$ , respectively, we have

$$\begin{aligned} u_i &= u_i(x_1, x_2, x_3; t) \quad (i=1,2,3), \\ w_i &= w_i(x_1, x_2, x_3; t) \quad (i=1,2,3), \end{aligned} \quad (2)$$

where  $x_i$  ( $i=1,2,3$ ) are the Cartesian coordinates of physical space. According to Lubensky *et al.*,<sup>17</sup> the phason represents relative motion of the constituent density waves, so it must be correlated with the physical space. The phonon-strain tensor is the spatial gradient of the phonon field while the phason-strain tensor is the spatial gradient of the phason field to the physical space, i.e.,

$$\begin{aligned} E_{ij} &= \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \quad (i, j=1,2,3), \\ W_{ij} &= \frac{\partial w_i}{\partial x_j} \quad (i, j=1,2,3). \end{aligned} \quad (3)$$

For the icosahedral quasicrystal, the generalized Hooke law<sup>25,26</sup> can be written as

$$T_{ij} = C_{ijkl} E_{kl} + R_{ijkl} W_{kl}, \quad H_{ij} = R_{ijkl} E_{kl} + K_{ijkl} W_{kl} \quad (4)$$

or

$$\begin{bmatrix} T \\ H \end{bmatrix} = \begin{bmatrix} [C] & [R] \\ [R^T] & [K] \end{bmatrix} \begin{bmatrix} E \\ W \end{bmatrix}, \quad (5)$$

with

$$C_{ijkl} = \lambda \delta_{ij} \delta_{kl} + \mu (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}), \quad (6)$$

$$[K] = \begin{bmatrix} K_1 & 0 & 0 & 0 & K_2 & 0 & 0 & K_2 & 0 \\ 0 & K_1 & 0 & 0 & -K_2 & 0 & 0 & K_2 & 0 \\ 0 & 0 & K_1 + K_2 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & K_1 - K_2 & 0 & K_2 & 0 & 0 & -K_2 \\ K_2 & -K_2 & 0 & 0 & K_1 - K_2 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & K_2 & 0 & K_1 & -K_2 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -K_2 & K_1 - K_2 & 0 & -K_2 \\ K_2 & K_2 & 0 & 0 & 0 & 0 & 0 & K_1 - K_2 & 0 \\ 0 & 0 & 0 & -K_2 & 0 & 0 & -K_2 & 0 & K_1 \end{bmatrix}, \quad (7)$$

and

$$[R]=R \begin{bmatrix} 1 & 1 & 1 & 0 & 0 & 0 & 0 & 1 & 0 \\ -1 & -1 & 1 & 0 & 0 & 0 & 0 & -1 & 0 \\ 0 & 0 & -2 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -1 & 1 & 0 & -1 \\ 1 & -1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & -1 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 & -1 & 1 & 0 & -1 \\ 1 & -1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & -1 & 0 & 0 & 1 \end{bmatrix}, \quad (8)$$

where  $T_{ij}$  and  $H_{ij}$  are the stresses corresponding to  $E_{ij}$  and  $W_{ij}$ , respectively,  $C_{ijkl}$  and  $K_{ijkl}$  the elastic constants in the phonon field and phason field, respectively, and  $R_{ijkl}$  the phonon-phason coupling elastic constants. Substituting Eqs. (6)–(8) into Eq. (5), we can rewrite the stress-strain relations as follows:

$$\begin{aligned} T_{11} &= \lambda \theta + 2\mu E_{11} + R(W_{11} + W_{22} + W_{33} + W_{13}), \\ T_{22} &= \lambda \theta - 2\mu E_{22} - R(W_{11} + W_{22} - W_{33} + W_{13}), \\ T_{33} &= \lambda \theta + 2\mu E_{33} - 2RW_{33}, \\ T_{23} &= 2\mu E_{23} + R(W_{32} - W_{12} - W_{21}) = T_{32}, \\ T_{31} &= 2\mu E_{31} + R(W_{11} - W_{22} + W_{31}) = T_{13}, \\ T_{12} &= 2\mu E_{12} + R(W_{21} - W_{23} - W_{12}) = T_{21}, \\ H_{11} &= R(E_{11} - E_{22} + 2E_{31}) + K_1 W_{11} + K_2(W_{31} + W_{13}), \\ H_{22} &= R(E_{11} - E_{22} - 2E_{31}) + K_1 W_{22} + K_2(W_{13} - W_{31}), \\ H_{33} &= R(E_{11} + E_{22} - 2E_{33}) + (K_1 + K_2)W_{33}, \\ H_{23} &= -2RE_{12} + (K_1 - K_2)W_{23} + K_2(W_{12} - W_{21}), \\ H_{31} &= 2RE_{31} + K_2(W_{11} - W_{22}) + (K_1 - K_2)W_{31}, \\ H_{12} &= -2R(E_{23} + E_{12}) + K_1 W_{12} + K_2(W_{23} + W_{32}), \\ H_{32} &= 2RE_{23} + (K_1 - K_2)W_{32} - K_2(W_{12} + W_{21}), \\ H_{13} &= R(E_{11} - E_{22}) + K_2(W_{11} + W_{22}) + (K_1 - K_2)W_{13}, \\ H_{12} &= 2R(E_{12} - E_{23}) - K_2(W_{23} + W_{32}) + K_1 W_{21}, \end{aligned} \quad (9)$$

where  $\theta = E_{11} + E_{22} + E_{33}$ . The wave equations are

$$\begin{aligned} \rho \frac{\partial^2 u_1}{\partial t^2} &= \frac{\partial T_{11}}{\partial x_1} + \frac{\partial T_{12}}{\partial x_2} + \frac{\partial T_{13}}{\partial x_3}, \\ \rho \frac{\partial^2 u_2}{\partial t^2} &= \frac{\partial T_{21}}{\partial x_1} + \frac{\partial T_{22}}{\partial x_2} + \frac{\partial T_{23}}{\partial x_3}, \end{aligned}$$

$$\rho \frac{\partial^2 u_3}{\partial t^2} = \frac{\partial T_{31}}{\partial x_1} + \frac{\partial T_{32}}{\partial x_2} + \frac{\partial T_{33}}{\partial x_3},$$

$$\rho \frac{\partial^2 w_1}{\partial t^2} = \frac{\partial H_{11}}{\partial x_1} + \frac{\partial H_{12}}{\partial x_2} + \frac{\partial H_{13}}{\partial x_3},$$

$$\rho \frac{\partial^2 w_2}{\partial t^2} = \frac{\partial H_{21}}{\partial x_1} + \frac{\partial H_{22}}{\partial x_2} + \frac{\partial H_{23}}{\partial x_3},$$

$$\rho \frac{\partial^2 w_3}{\partial t^2} = \frac{\partial H_{31}}{\partial x_1} + \frac{\partial H_{32}}{\partial x_2} + \frac{\partial H_{33}}{\partial x_3}, \quad (10)$$

where  $\rho$  is the mean mass density of the quasicrystal. Assuming that the propagation of vibration is in the direction  $\vec{\varphi}$  of physical subspace and using  $l, m$ , and  $n$  as the direction cosines of  $\vec{\varphi}$ , we have

$$\varphi = lx_1 + mx_2 + nx_3, \quad (11)$$

where  $\varphi$  is the length of  $\vec{\varphi}$ . Then Eq. (3) becomes

$$\begin{aligned} E_{11} &= l \frac{\partial u_1}{\partial \varphi}, \quad E_{22} = m \frac{\partial u_2}{\partial \varphi}, \quad E_{33} = n \frac{\partial u_3}{\partial \varphi}, \\ 2E_{23} &= m \frac{\partial u_3}{\partial \varphi} + n \frac{\partial u_2}{\partial \varphi}, \quad 2E_{13} = \left( n \frac{\partial u_1}{\partial \varphi} + l \frac{\partial u_3}{\partial \varphi} \right), \\ 2E_{12} &= m \frac{\partial u_1}{\partial \varphi} + l \frac{\partial u_2}{\partial \varphi}, \quad W_{11} = l \frac{\partial w_1}{\partial \varphi}, \quad W_{22} = m \frac{\partial w_2}{\partial \varphi}, \\ W_{33} &= n \frac{\partial w_3}{\partial \varphi}, \quad W_{23} = n \frac{\partial w_2}{\partial \varphi}, \quad W_{31} = l \frac{\partial w_3}{\partial \varphi}, \\ W_{12} &= m \frac{\partial w_1}{\partial \varphi}, \quad W_{32} = m \frac{\partial w_3}{\partial \varphi}, \quad W_{13} = n \frac{\partial w_1}{\partial \varphi}, \\ W_{21} &= l \frac{\partial w_2}{\partial \varphi}. \end{aligned} \quad (12)$$

Substituting Eq. (12) into Eq. (9), and then into Eq. (10), we can obtain

$$\begin{aligned}
 \rho \frac{\partial^2 u_1}{\partial t^2} &= \Gamma_{11} \frac{\partial^2 u_1}{\partial \varphi^2} + \Gamma_{12} \frac{\partial^2 u_2}{\partial \varphi^2} + \Gamma_{13} \frac{\partial^2 u_3}{\partial \varphi^2} + \Gamma_{14} \frac{\partial^2 w_1}{\partial \varphi^2} \\
 &\quad + \Gamma_{15} \frac{\partial^2 w_2}{\partial \varphi^2} + \Gamma_{16} \frac{\partial^2 w_3}{\partial \varphi^2}, \\
 \rho \frac{\partial^2 u_2}{\partial t^2} &= \Gamma_{21} \frac{\partial^2 u_1}{\partial \varphi^2} + \Gamma_{22} \frac{\partial^2 u_2}{\partial \varphi^2} + \Gamma_{23} \frac{\partial^2 u_3}{\partial \varphi^2} + \Gamma_{24} \frac{\partial^2 w_1}{\partial \varphi^2} \\
 &\quad + \Gamma_{25} \frac{\partial^2 w_2}{\partial \varphi^2} + \Gamma_{26} \frac{\partial^2 w_3}{\partial \varphi^2}, \\
 \rho \frac{\partial^2 u_3}{\partial t^2} &= \Gamma_{31} \frac{\partial^2 u_1}{\partial \varphi^2} + \Gamma_{32} \frac{\partial^2 u_2}{\partial \varphi^2} + \Gamma_{33} \frac{\partial^2 u_3}{\partial \varphi^2} + \Gamma_{34} \frac{\partial^2 w_1}{\partial \varphi^2} \\
 &\quad + \Gamma_{35} \frac{\partial^2 w_2}{\partial \varphi^2} + \Gamma_{36} \frac{\partial^2 w_3}{\partial \varphi^2}, \\
 \rho \frac{\partial^2 w_1}{\partial t^2} &= \Gamma_{41} \frac{\partial^2 u_1}{\partial \varphi^2} + \Gamma_{42} \frac{\partial^2 u_2}{\partial \varphi^2} + \Gamma_{43} \frac{\partial^2 u_3}{\partial \varphi^2} + \Gamma_{44} \frac{\partial^2 w_1}{\partial \varphi^2} \\
 &\quad + \Gamma_{45} \frac{\partial^2 w_2}{\partial \varphi^2} + \Gamma_{46} \frac{\partial^2 w_3}{\partial \varphi^2}, \\
 \rho \frac{\partial^2 w_2}{\partial t^2} &= \Gamma_{51} \frac{\partial^2 u_1}{\partial \varphi^2} + \Gamma_{52} \frac{\partial^2 u_2}{\partial \varphi^2} + \Gamma_{53} \frac{\partial^2 u_3}{\partial \varphi^2} + \Gamma_{54} \frac{\partial^2 w_1}{\partial \varphi^2} \\
 &\quad + \Gamma_{55} \frac{\partial^2 w_2}{\partial \varphi^2} + \Gamma_{56} \frac{\partial^2 w_3}{\partial \varphi^2}, \\
 \rho \frac{\partial^2 w_3}{\partial t^2} &= \Gamma_{61} \frac{\partial^2 u_1}{\partial \varphi^2} + \Gamma_{62} \frac{\partial^2 u_2}{\partial \varphi^2} + \Gamma_{63} \frac{\partial^2 u_3}{\partial \varphi^2} + \Gamma_{64} \frac{\partial^2 w_1}{\partial \varphi^2} \\
 &\quad + \Gamma_{65} \frac{\partial^2 w_2}{\partial \varphi^2} + \Gamma_{66} \frac{\partial^2 w_3}{\partial \varphi^2}, \tag{13}
 \end{aligned}$$

where

$$\begin{aligned}
 \Gamma_{11} &= \lambda l^2 + \mu m^2 + \mu n^2 + 2\mu l^2, & \Gamma_{21} &= (\lambda + \mu)lm, \\
 \Gamma_{12} &= (\lambda + \mu)lm, & \Gamma_{22} &= \lambda m^2 + \mu l^2 + \mu n^2 - 2\mu m^2, \\
 \Gamma_{13} &= (\lambda + \mu)ln, & \Gamma_{23} &= (\lambda + \mu)mn, \\
 \Gamma_{14} &= Rl^2 - Rm^2 + 2Rln, & \Gamma_{24} &= -2Rlm - 2Rmn, \\
 \Gamma_{15} &= 2Rlm - 2Rmn, & \Gamma_{25} &= -2Rm^2 - 2Rln, \\
 \Gamma_{16} &= 2Rln, & \Gamma_{26} &= 2Rmn,
 \end{aligned} \tag{14}$$

$$\begin{aligned}
 \Gamma_{31} &= (\lambda + \mu)ln, & \Gamma_{41} &= Rl^2 - Rm^2 + 2Rln, \\
 \Gamma_{32} &= (\lambda + \mu)mn, & \Gamma_{42} &= 2Rlm - 2Rmn, \\
 \Gamma_{33} &= \lambda n^2 + \mu l^2 + \mu m^2 + 2\mu n^2, & \Gamma_{43} &= Rl^2 - Rm^2, \\
 \Gamma_{34} &= Rl^2 - Rm^2, \\
 \Gamma_{44} &= K_1 l^2 + K_1 m^2 + (K_1 - K_2)n^2 + 2K_2 ln, \\
 \Gamma_{35} &= -2Rlm, & \Gamma_{45} &= 2K_2 mn, \\
 \Gamma_{36} &= Rl^2 + Rm^2 - 2Rn^2, & \Gamma_{46} &= K_2 l^2 + K_2 m^2, \\
 \Gamma_{51} &= 2Rlm - 2Rmn, & \Gamma_{61} &= 2Rln, \\
 \Gamma_{52} &= Rl^2 - Rm^2 - 2Rln, & \Gamma_{62} &= 2Rmn, \\
 \Gamma_{53} &= -2Rlm, & \Gamma_{63} &= Rl^2 + Rm^2 - 2Rn^2, \\
 \Gamma_{54} &= 2K_2 mn, & \Gamma_{64} &= K_2 l^2 - K_2 m^2, \\
 \Gamma_{55} &= K_1(l^2 + m^2) + (K_1 - K_2)n^2 - 2K_2 ln, & \Gamma_{65} &= -2K_2 lm, \\
 \Gamma_{56} &= 2K_2 lm, \\
 \Gamma_{66} &= (K_1 - K_2)l^2 + (K_1 - K_2)m^2 + (K_1 + K_2)n^2.
 \end{aligned} \tag{15}$$

With  $\vec{\xi}$  denoting elastic displacement vector resulting from the wave propagation along the  $\vec{\varphi}$  direction and  $p, q, r, p', q', r'$  denoting its direction cosines in the six-dimensional superspace, we have

$$\begin{aligned}
 u_1 &= p\xi, & u_2 &= q\xi, & u_3 &= r\xi, & w_1 &= p'\xi, \\
 w_2 &= q'\xi, & w_3 &= r'\xi, \\
 \xi &= pu_1 + qu_2 + ru_3 + p'w_1 + q'w_2 + r'w_3, \tag{17}
 \end{aligned}$$

where  $\xi$  is the length of  $\vec{\xi}$ . Substituting Eq. (17) into Eq. (13), the latter can be reduced to the wave equation

$$\rho \frac{\partial^2 \xi}{\partial t^2} = C^* \frac{\partial^2 \xi}{\partial \varphi^2}, \tag{18}$$

where  $C^*$  is the effective elastic constant and satisfies the following equations:

$$\begin{aligned}
 p\Gamma_{11} + q\Gamma_{12} + r\Gamma_{13} + p'\Gamma_{14} + q'\Gamma_{15} + r'\Gamma_{16} &= pC^*, \\
 p\Gamma_{21} + q\Gamma_{22} + r\Gamma_{23} + p'\Gamma_{24} + q'\Gamma_{25} + r'\Gamma_{26} &= qC^*, \\
 p\Gamma_{31} + q\Gamma_{32} + r\Gamma_{33} + p'\Gamma_{34} + q'\Gamma_{35} + r'\Gamma_{36} &= rC^*, \\
 p\Gamma_{41} + q\Gamma_{42} + r\Gamma_{43} + p'\Gamma_{44} + q'\Gamma_{45} + r'\Gamma_{46} &= p'C^*, \\
 p\Gamma_{51} + q\Gamma_{52} + r\Gamma_{53} + p'\Gamma_{54} + q'\Gamma_{55} + r'\Gamma_{56} &= q'C^*, \\
 p\Gamma_{61} + q\Gamma_{62} + r\Gamma_{63} + p'\Gamma_{64} + q'\Gamma_{65} + r'\Gamma_{66} &= r'C^*. \tag{19}
 \end{aligned}$$

The condition that Eq. (19) have a solution leads to

$$\begin{pmatrix} \Gamma_{11}-C^* & \Gamma_{12} & \Gamma_{13} & \Gamma_{14} & \Gamma_{15} & \Gamma_{16} \\ \Gamma_{21} & \Gamma_{22}-C^* & \Gamma_{23} & \Gamma_{24} & \Gamma_{25} & \Gamma_{26} \\ \Gamma_{31} & \Gamma_{32} & \Gamma_{33}-C^* & \Gamma_{34} & \Gamma_{35} & \Gamma_{36} \\ \Gamma_{41} & \Gamma_{42} & \Gamma_{43} & \Gamma_{44}-C^* & \Gamma_{45} & \Gamma_{46} \\ \Gamma_{51} & \Gamma_{52} & \Gamma_{53} & \Gamma_{54} & \Gamma_{55}-C^* & \Gamma_{56} \\ \Gamma_{61} & \Gamma_{62} & \Gamma_{63} & \Gamma_{64} & \Gamma_{65} & \Gamma_{66}-C^* \end{pmatrix} = 0. \quad (20)$$

To calculate the effective elastic constant, we must first get the values of  $\Gamma_{ij}$ . However,  $\Gamma_{ij}$  are determined by the elastic constant and direction cosine of the propagating direction, so we first calculate the direction cosine by the projection method. According to the result of Elser,<sup>23</sup> we have

$$(h_1 h_2 h_3 h_4 h_5 h_6) \tilde{Q}_{\parallel}^{-1} = (x_1, x_2, x_3), \quad (21)$$

with

$$\tilde{Q}_{\parallel}^{-1} = a \begin{pmatrix} \tau & 0 & 1 \\ \tau & 0 & -1 \\ 0 & 1 & -\tau \\ -1 & \tau & 0 \\ 0 & 1 & \tau \\ 1 & \tau & 0 \end{pmatrix}, \quad (22)$$

where  $h_1, h_2, h_3, h_4, h_5, h_6$  and  $x_1, x_2, x_3$  are coordinates based on the basic vector  $d_{\parallel}^i$  ( $i=1, 2, \dots, 6$ ) of  $V_{\parallel}$  of six-dimensional superspace and Cartesian coordinates of  $V_{\parallel}$ , respectively;  $a=1/\sqrt{2(1+\tau^2)}$ . Thus the direction cosine  $(l, m, n)$  of physical space can be written as

$$l = \frac{x_1}{\sqrt{x_1^2 + x_2^2 + x_3^2}}, \quad m = \frac{x_2}{\sqrt{x_1^2 + x_2^2 + x_3^2}},$$

$$n = \frac{x_3}{\sqrt{x_1^2 + x_2^2 + x_3^2}}. \quad (23)$$

Combining Eqs. (14)–(16) we can obtain the solution of Eq. (20) and then calculate the phase velocities of wave propagation along any direction.

### III. LOW-TEMPERATURE LATTICE EXCITATION OF THE ICOSAHEDRAL $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$ QUASICRYSTAL

#### A. Phase velocities of phonons and phasons

We now concentrate on investigating the phonon and phason velocities in the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal along the fivefold  $(1, 0, 0, 0, 0)$ , twofold  $(1, -1, 0, 0, 0)$ , and threefold  $(1, 1, -1, 1, -1)$  directions, respectively. Here the experimentally determined data  $\rho=5.1 \text{ g cm}^{-3}$  was used<sup>27</sup> and the low-temperature values  $\lambda=0.75$ ,  $\mu=0.65$  ( $10^{12} \text{ dyn/cm}^2$ ) of the elastic moduli  $C_{ij}$  obtained using resonant ultrasound spectroscopy were taken.<sup>28</sup>  $K_1=0.8\sim 1.1$  is cal-

culated with the transfer-matrix method by Newman and Henley;<sup>29</sup>  $K_2/K_1=-0.60\pm 0.02$  obtained by Capitan *et al.* with x-ray diffuse scattering from the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal.<sup>30</sup> In this paper, we choose  $K_1=0.81$ ,  $K_2=-0.50$  ( $10^{12} \text{ dyn/cm}^2$ ). On the phonon-phason coupling constant, Zhu and Henley<sup>31</sup> proved its magnitude relative to the phonon and phason elastic constants of order 1/10 and calculated  $R=0.0066$  ( $10^{12} \text{ dyn/cm}^2$ ) for the Al-Mn-Pd quasicrystal. For the fivefold axis  $(1, 0, 0, 0, 0)$  direction, substituting  $h_1=1$  and  $h_2=h_3=h_4=h_5=h_6=0$  into Eq. (21) and then into Eq. (23), we have  $l=\tau/(\sqrt{1+\tau^2})$ ,  $m=0$ ,  $n=1/(\sqrt{1+\tau^2})$ , where  $\tau=(1+\sqrt{5})/2$  is the golden mean of the Fibonacci lattice. Further, inserting the values of  $l, m, n$  and  $C_{ij}, K_1, K_2$  into Eqs. (14)–(16) and then into Eq. (20), we can work out the effective elastic modulus  $C_i^*$  ( $i=1, 2, \dots, 6$ ). According to  $v_i=\sqrt{C_i^*/\rho}$ , we have  $v_1=6352.28$ ,  $v_2=v_3=3576.71$ ,  $v_4=2501.56$ ,  $v_5=5240.84$ , and  $v_6=4893.35$  ( $m/s$ ), where  $v_i$  ( $i=1, \dots, 6$ ) are the six phase velocities of wave propagation along the  $(1, 0, 0, 0, 0)$  direction of the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal. The  $v_1$  stands for a phase velocity of the longitudinal acoustic phonon mode;  $v_2$  and  $v_3$  are phase velocities of two transversal acoustic phonon modes;  $v_4, v_5, v_6$  are the phase velocities of three acoustic phason modes. Similarly, we can calculate the phase velocities of waves propagating along the twofold  $(1, -1, 0, 0, 0)$  and threefold  $(1, 1, -1, 1, -1)$  directions and have listed them in Table I, where the coefficient parameter  $\eta$  is taken from Ref. 15. From Table I, we note that  $v_i$  ( $i=1, 2, 3$ ) of our theoretical results are in good agreement with the results of the resonant ultrasound spectroscopy experiment.<sup>27</sup> In addition, we also find from Table I that the phase velocities of acoustic phonons and phasons possess small anisotropy. This might be the reason that the icosahedral quasicrystals have the small anisotropy of the temperature dependence of the magnetoresistivity.<sup>5</sup>

#### B. Density of vibrational states

Debye<sup>32</sup> regarded a crystal as a continuous elastic medium. We now extend the Debye hypothesis to quasicrystals. That is, we can regard an icosahedral quasicrystal as a continuous elastic medium. With  $\omega$  denoting the atom vibrational angular frequency and  $g(\omega)$  denoting the frequency distribution function, i.e., the density of vibrational states, we have, by following the argument of Walti *et al.*,<sup>15</sup>

TABLE I. Coefficient parameters and velocities of the acoustic phonons and phasons of the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal in the range  $|q| < 0.35 \text{ \AA}^{-1}$ .

Axis	Direction	$v_1$ (m/s)	$\eta \times 10^{-17}$ (m <sup>3</sup> /s)	$v_2$ (m/s)	$v_3$ (m/s)	$v_4$ (m/s)	$v_5$ (m/s)	$v_6$ (m/s)
5	(1,0,0,0,0)	6352.3	-3.9	3576.9	3576.9	2501.6	5240.8	4893.4
2	(1,-1,0,0,0,0)	6340.0	-3.9	3570.0	3570.0	2465.5	5068.2	5068.2
3	(1,1,-1,1,1,-1)	6352.6	-3.9	3577.1	3577.1	2508.7	5256.0	4874.4

$$g(\omega) = \begin{cases} a\omega^2 + b\omega^4 & \text{for } \omega \leq \omega_0, \\ 0 & \text{for } \omega > \omega_0. \end{cases} \quad (24)$$

We should note that the phason modes, although they do exist, do not create new degrees of freedom, so the total freedom number remains 3 times the number of particles;<sup>1</sup> i.e., there are  $3N_A$  simple harmonic vibrations in the quasicrystal, where  $N_A$  stands for the number of atoms in 1 mol icosahedral quasicrystal, i.e.,

$$\int_0^\infty g(\omega) d\omega = 3N_A. \quad (25)$$

The substitution of Eq. (24) into Eq. (25) leads to

$$3N_A = \frac{a}{3}\omega_0^3 + \frac{b}{5}\omega_0^5. \quad (26)$$

Based on the assumption of phason-propagating modes, the dispersion relation of acoustic excitations  $\omega(q)$  can be expanded in a power series of the form

$$\omega_i(q) = v_i q + \eta_i q^3 + O(q^5) \quad (i = 1, \dots, 6), \quad (27)$$

where the parameter  $v_i$  and  $\eta_i$  data in the wave vector range  $|q| < 0.35 \text{ \AA}^{-1}$  are given in Table I. In the acoustic limit, the dispersion relation  $\omega(q)$  for the quasicrystal with an icosahedral point-group symmetry depends only on the absolute value of  $\mathbf{q}$ , leading to a DOVS of the acoustic modes of the form described by Eq. (24), where the coefficient parameters satisfy

$$a = \frac{3M}{2\pi^2\rho} \frac{1}{6} \sum_{i=1}^6 \frac{1}{v_i^3}, \quad b = -\frac{15M}{2\pi^2\rho} \frac{1}{6} \sum_{i=1}^6 \frac{\eta_i}{v_i^6}, \quad (28)$$

where  $M$  and  $\rho$  represent the molar mass and mass density of the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal, respectively. We note that the coefficients  $a$  and  $b$  contain contributions from both the phonons and phasons since six phase velocities are involved. According to neutron-scattering experiments for other icosahedral quasicrystals, the parameters  $\eta_i$  ( $i = 1, 2, 3$ ) are of comparable magnitude.<sup>15</sup> Therefore, we take  $\eta_i = \eta$ . For the phasons, we also assume that  $\eta_i = \eta$  ( $i = 4, 5, 6$ ). Substituting the velocities and  $\eta$  parameters of Table I into the Eq. (28), we obtain  $a = 3.00 \times 10^{-17} \text{ s}^3/\text{rad}^3 \text{ mol}$ ,  $b = 2.38 \times 10^{-43} \text{ s}^5/\text{rad}^5 \text{ mol}$  which are close to the experimental data:  $a = 3.27 \times 10^{-17} \text{ s}^3/\text{rad}^3 \text{ mol}$ ,  $b = 2.37 \times 10^{-43} \text{ s}^5/\text{rad}^5 \text{ mol}$  measured by Wälti *et al.*<sup>15</sup> However, their theoretical values<sup>15</sup>  $a = 1.8 \times 10^{-17} \text{ s}^3/\text{rad}^3 \text{ mol}$ ,  $b = 0.65 \times 10^{-43} \text{ s}^5/\text{rad}^5 \text{ mol}$  are distinctly smaller than their experimental data. Comparing our theoretical formula with those used by Wälti *et al.*, we can see that the difference is mainly caused by their neglecting the contribution of phasons to the DOVS. Inserting our results of  $a, b$  into Eq. (26), we have  $\omega_0 = 3.155 \times 10^{15} \text{ rad/s}$ .

### C. Lattice specific heat

In the harmonic approximation, the lattice specific heat  $C_{ph}(T)$  depends on the DOVS  $g(\omega)$ :

 TABLE II. The coefficients of the DOVS and  $C_{ph}$  of the icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal obtained from the experimental measurement (Ref. 15) and theoretical calculation, where the theoretical value 1 (theor. 1) (Ref. 15) represents the results only containing the contribution of acoustic phonons. The theoretical values 2–4 represent the results containing the contributions of acoustic phonons, acoustic phasons, and phonon-phason coupling to the DOVS and lattice specific heat by using the phase velocities along the fivefold, twofold, and threefold axis directions, respectively.

	DOVS		$C_{ph}$		$\Theta_D(\text{K})$
	$a(\text{s}^3/\text{rad}^3 \text{ mol})$	$b(\text{s}^5/\text{rad}^5 \text{ mol})$	$\beta(\text{J/mol K}^4)$	$\delta(\text{J/mol K}^6)$	
Expt. data <sup>a</sup>	$3.27 \times 10^{-17}$	$2.37 \times 10^{-43}$	$2.63 \times 10^{-5}$	$9.2 \times 10^{-8}$	420
Theor. 1	$1.8 \times 10^{-17}$	$0.65 \times 10^{-43}$	$1.63 \times 10^{-5}$	$2.5 \times 10^{-8}$	492
Theor. 2	$3.00 \times 10^{-17}$	$2.38 \times 10^{-43}$	$2.43 \times 10^{-5}$	$9.27 \times 10^{-8}$	431
Theor. 3	$3.07 \times 10^{-17}$	$2.56 \times 10^{-43}$	$2.44 \times 10^{-5}$	$9.96 \times 10^{-8}$	430
Theor. 4	$3.01 \times 10^{-17}$	$2.38 \times 10^{-43}$	$2.43 \times 10^{-5}$	$9.26 \times 10^{-8}$	431

<sup>a</sup>Reference 15.

$$C_{ph} = \int_0^\infty g(\omega) k_B \frac{(\hbar \omega / k_B T)^2 \exp(\hbar \omega / k_B T)}{[\exp(\hbar \omega / k_B T) - 1]^2} d\omega. \quad (29)$$

Substituting Eq. (24) into Eq. (29), then integrating Eq. (29) with considering Eq. (26), (28), we obtain

$$C_{ph} = \beta T^3 + \delta T^5, \quad (30)$$

where

$$\beta = \left[ 9N_A \left( \frac{k_B}{\hbar \omega_0} \right)^3 - \frac{3b}{5} \omega_0^2 \left( \frac{k_B}{\hbar} \right)^3 \right] \frac{4k_B \pi^4}{15} \quad (31)$$

and

$$\delta = 6\hbar b \left( \frac{k_B}{\hbar} \right)^6 \frac{8\pi^6}{63}. \quad (32)$$

Inserting our calculated  $a, b$ , and  $\omega_0$  into Eqs. (31) and (32), we have  $\beta = 2.43 \times 10^{-5} \text{ J mol}^{-1} \text{ K}^{-4}$  and  $\delta = 9.27 \times 10^{-8} \text{ J mol}^{-1} \text{ K}^{-6}$ , which corresponds to the Debye temperature 430 K.

#### IV. DISCUSSION AND CONCLUSION

In order to further compare our theoretical results with the experimental data, we have calculated the coefficient parameters of the DOVS and lattice specific-heat expression by using the phase velocities along the fivefold, twofold, and threefold directions, respectively, and listed them in Table II. From Table II, we can see that all of the theoretical results containing the contributions of phonons and phasons and the phonon-phason coupling are in agreement with the experimental data, in which the coefficient of the  $\omega^2$  or  $T^3$  term is a little smaller than the experimental data but that of the  $\omega^4$  or  $T^5$  term is almost equal to the experimental data. However, the theoretical values<sup>15</sup> neglecting the contribution of the phason are much smaller than the experimental data, in which the coefficient of the  $\omega^2$  or  $T^3$  term is almost half of the experimental data, as well as that of the  $\omega^4$  or  $T^5$  term being about one-fifth of the experimental data. These results show that the contribution of phasons cannot be neglected. The higher the order of the DOVS or specific-heat term is, the stronger the affection is.

Now, we show a double logarithmic plot of  $C_{ph}/T^3$  vs  $T$  in Fig. 1. The solid circles represent the experimental curve (EC) measured by Wälti *et al.*,<sup>15</sup> the open triangles (PPC) are our theoretical results including the contributions of the acoustic phonons and phasons and the phonon-phason coupling to the lattice specific heat, and the open squares (PC) are the theoretical results only considering the contribution of the acoustic phonons. It is clear from Fig. 1 that the values of the PPC curve are in agreement with the experimental data measured by Wälti *et al.* below 15 K, but those of the PC curve are distinctly smaller than the experimental data. Above 15 K, the EC curve increases slowly with increasing temperature and passes over a broad maximum centered at approximately 25 K, and then starts to decrease. The PPC curve goes up a little faster than the EC one. For this difference the first possible reason would be that the density of

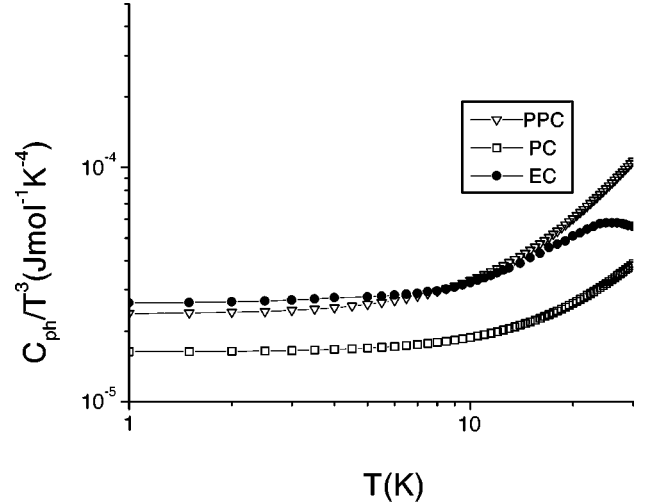


FIG. 1. The lattice specific heat of  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  is plotted as  $C_{ph}/T^3$  vs  $T$ . The solid circles exhibit the experimental results, the open triangles the theoretical values containing the contributions of the acoustic phonons, phasons, and phonon-phason coupling, and the open squares the theoretical results only containing the contribution of acoustic phonons.

states would no longer increase by the rule  $a\omega^2 + b\omega^4$  but approach a constant, and then decrease to zero<sup>33</sup> with temperature increase. The second one might be that the phonon excitation modes would increase so that the accuracy of the continuous elastic model would decrease.

#### V. SUMMARY

In this paper, we first suggest a method to deal with the contributions of phasons and phonon-phason coupling to the DOVS and lattice specific heat of quasicrystals at low temperature. We have calculated the six phase velocities of waves propagating in the icosahedral quasicrystal. By adding the phason phase velocities to the average velocity expression, we succeed in explaining the DOVS and lattice specific-heat experiments performed on an icosahedral  $\text{Al}_{68.2}\text{Mn}_9\text{Pd}_{22.8}$  quasicrystal below 15 K. Our results show that the contribution of the phasons to the DOVS and specific heat cannot be neglected at low temperature. The consistency of our theoretical results with the experimental data demonstrates that our equivalent assumption of phonons and phasons would be plausible for dealing with the vibrational properties of quasicrystals at low temperature. Even though in the present stage this is a phenomenological treatment, we believe that it might reveal the intrinsic properties of quasicrystals. Thus, we believe that our theory should be helpful to explain the thermal, electrical, and optical properties which are related to the phasons of quasicrystals at low temperature.

#### ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China, Grant No. 19874021.

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