

Application of the Debye model to study anharmonic correlation effects for the CuAgX (X = 72; 50) intermetallic alloy

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Abstract

Based on the Debye - Waller factor in extended x-ray absorption fine structure spectra, the correlated displacement function, mean square displacement (MSD) and mean square relative displacement (MSRD) were determined. Analytical expressions of MSD and MSRD were considered using Debye models. Challenging problems due to many-particles effects were replaced by a calculation based on the anharmonic effective potential, including the interaction of absorbing and scattering atoms with their nearest neighbours atoms. The difference between MSRD and MRD was analyzed. The methods were applied to face-centred-cubic crystals and their alloys, and have discovered irregularities in the structure of CuAg (at a 50:50 rate) alloy at low temperatures. Numerical results for copper (Cu) crystal and copper-silver alloys (CuAgX (X = 72; 50)) agreed with experimental values and calculations conducted in other studies.

Keywords: Debye-Waller factor, anharmonic effective potential, CuAgX (X = 72; 50), anharmonic correlation, displacement correlation function

(Some figures may appear in colour only in the online journal)

1. Introduction

The thermal oscillations in Extended x-ray Absorption Fine Structure (EXAFS) that give rise to Debye-Waller factors (DWF) $e^{-W(T)}$ have been considered in previous studies [1, 2], and it has been found that the DWF is related to the damping of the EXAFS amplitude as the temperature increases. Based on the expansion of the cumulants due to thermal vibrations of atoms, the $W(T)$ is reduced in an exponential function to $W(T) \approx 2k^2\sigma^2(T)$, where $\sigma^2(T)$ is the Mean Square Relative Displacement (MSRD) of the bond between absorbing and backscattering atoms [3]. Similarly, during the diffraction of neutrons or x-ray absorption, the DWF has the form $W(T) = (1/2)k^2u^2(T)$. However, the DWF in EXAFS spectra refers to correlated averages over the relative displacement of $\sigma^2(T)$ for a pair of absorber and backscatter atoms. In contrast, neutron diffraction $u^2(T)$ refers to the Mean Square Displacement (MSD) of a given atom. The functions $\sigma^2(T)$ and $u^2(T)$ are closely related to one another, and from them, the Displacement-Displacement Correlation Function

(DDCF) $C_R(T)$ can be deduced to describe the correlation effects in the vibration of atoms. The DWF has an essential role in the determination of crystal structures as well as thermal quantities in the EXAFS spectra. Many efforts have been made to derive the procedures for the calculation and analysis of $\sigma^2(T)$ [1, 2, 4, 3] and $u^2(T)$ [5, 6]. However, correlation effects for intermetallic alloys have not mentioned by many studies.

The purpose of this work is to study the correlation effects in atomic vibrations of cubic crystals in XAFS, i.e., to develop a new procedure for calculation of the $C_R(T)$ for atomic vibration in the cubic crystals (fcc) and their alloys in XAFS. Expression for the MSD $u^2(T)$ has been derived. Using it and the MSRD $\sigma^2(T)$ we derive $C_R(T)$. The effective interaction potential of the system has been considered by taking into account the influences of the nearest atomic neighbors based on the anharmonic correlated Debye model. This potential contains the Morse potential characterizing the interaction of each pair of atoms. The difference between the $\sigma^2(T)$ obtained from the ACDM and $u^2(T)$ from the ADM

was analyzed. The numerical results for application to copper (Cu) crystal and CuAgX (X = 72; 50) alloys. The expression CuAgX (X = 72) refers to an alloy with 72% Cu and 28% Ag ratio, and CuAgX (X = 50) refers to an alloy with Cu:Ag in a 50:50 ratio. These materials have studied by authors in some works [7, 8], and have discovered irregularities in the structure of CuAg (at 50:50 rate) alloy at low temperatures, accordingly, Cu-Ag alloys do not form an ordered phase at a molar composition of 1:1. The results obtained by the present theory have been compared and agree well with experimental values [9–119-11] and other studies [12, 8, 6].

2. Formalism

In the EXAFS theory, the quantity $\sigma^2(T)$ is defined while taking the exponential averages $\exp(2ik.r_j)$ in the form [2]

$$\langle \exp(2ik.r_j) \rangle \rightarrow \langle \exp(2ik\Delta_j) \rangle = \exp(-2k^2\langle \Delta_j^2 \rangle). \quad (1)$$

The factor of expression (1) determines the damping coefficient due to thermal vibrations in the EXAFS spectra. It is also known as the Debye–Waller factor or the MSRD. In expression (1), $\Delta_j = \hat{\mathbf{R}}_j^0 \cdot (\mathbf{u}_j - \mathbf{u}_0)$, where $\hat{\mathbf{R}}_j^0$ is a unit vector for atom j at equilibrium, \mathbf{u}_j is a displacement vector of atom j , and \mathbf{u}_0 is the displacement vector of the absorbing atom located at the coordinate origin. For the harmonic approximation oscillation,

$$\sigma_j^2 = \langle \Delta_j^2 \rangle. \quad (2)$$

Substitute $\Delta_j = \hat{\mathbf{R}}_j^0 \cdot (\mathbf{u}_j - \mathbf{u}_0)$ into expression (2), MSRD $\sigma^2(T)$ have the form:

$$\sigma_j^2(T) = \langle [\hat{\mathbf{R}}_j \cdot (\mathbf{u}_j - \mathbf{u}_0)]^2 \rangle = \langle (\mathbf{u}_j \cdot \hat{\mathbf{R}}_j)^2 \rangle + \langle (\mathbf{u}_0 \cdot \hat{\mathbf{R}}_j)^2 \rangle - 2\langle (\mathbf{u}_0 \cdot \hat{\mathbf{R}}_j)(\mathbf{u}_j \cdot \hat{\mathbf{R}}_j) \rangle. \quad (3)$$

With $\mathbf{u}_0 = \mathbf{u}_j$, we have the mean square displacement (MSD):

$$u_j^2(T) = \langle (\mathbf{u}_0 \cdot \hat{\mathbf{R}}_j)^2 \rangle = \langle (\mathbf{u}_j \cdot \hat{\mathbf{R}}_j)^2 \rangle, \quad (4)$$

and the correlated function DDCF:

$$C_R(T) = 2\langle (\mathbf{u}_0 \cdot \hat{\mathbf{R}}_j)(\mathbf{u}_i \cdot \hat{\mathbf{R}}_j) \rangle. \quad (5)$$

From expressions (3), (4), (5), the relation expression can be deduced:

$$\sigma_j^2(T) = 2u_j^2(T) - C_R(T). \quad (6)$$

To consider anharmonic contributions to the MSRD $\sigma_j^2(T)$ due to temperature, hence the temperature dependence for the correlation function DDCF $C_R(T)$ and MSD $u^2(T)$, we used an argument for its change due to the temperature increase and obtain $\sigma^2(T) - \sigma^2(T_0) = (1 + \beta(T))[\sigma_H^2 - \sigma^2(T_0)]$, with $\beta = 2\gamma_G\Delta V/V$; γ_G is Gruneisen parameter, and $\Delta V/V$ is the relative volume change due to thermal expansion, T_0 is a very low temperature so that $\sigma^2(T_0)$ is harmonic MSRD. Developing further, we obtain the total MSRD $\sigma^2(T) = \sigma_H^2(T) + \beta(T)[\sigma_H^2 - \sigma^2(T_0)]$ (*). It is clear that the MSRD approaches the very small value of zero-point contribution σ_0^2 when the temperature approaches zero, i.e.,

$\sigma^2(T_0) \rightarrow \sigma_0^2$ for $T_0 \rightarrow 0$. Hence, it can be seen in expression (*) that the total MSRD $\sigma^2(T)$ at a given temperature T consists of the harmonic contribution $\sigma_H^2(T)$ and the anharmonic one $\sigma_A^2(T)$, $\sigma^2(T) = \sigma_H^2(T) + \sigma_A^2(T)$; and $\sigma_A^2(T) = \beta(T)[\sigma_H^2 - \sigma_0^2]$.

For harmonic approximation, at low temperatures, $\sigma_j^2(T)$ is isotropic and shows Gaussian symmetry. When the temperature rises to a certain critical value, the interaction between atoms will be asymmetric, anharmonic effects will appear, and the phonon interaction must be taken into account. To determine thermodynamic parameters with an anharmonic effect, we need to determine the effective elastic force constants (force constants) of atomic pairs in a cluster. The determination of force constants is based on the effective anharmonic potential as a function along the direction of the displacement x . According to ACDM, the anharmonic potential has the following form [3, 13]:

$$V_{\text{eff}}^A(x) \approx \frac{1}{2}k_{\text{eff}}^A x^2 + k_{3\text{eff}}^A x^3, \quad (7)$$

and in the ADM, the anharmonic potential has the form [14]

$$V_{\text{eff}}^D(x) \approx \frac{1}{2}k_{\text{eff}}^D x^2 + k_{3\text{eff}}^D x^3. \quad (8)$$

Here, k_{eff}^A and $k_{3\text{eff}}^D$ are effective elastic force constants, $k_{3\text{eff}}^A$ and $k_{3\text{eff}}^D$ are cubic parameters that cause asymmetry of the interaction potential due to anharmonicity, x is the lattice thermal expansion. The difference between the $V_{\text{eff}}^A(x)$ and $V_{\text{eff}}^D(x)$ potentials is due to the difference between force constants k_{eff}^A and k_{eff}^D and the difference between cubic parameters $k_{3\text{eff}}^A$ and $k_{3\text{eff}}^D$.

The values of the force constant k_{eff}^A and the cubic parameter $k_{3\text{eff}}^A$ can be obtained when $V_{\text{eff}}^A(x)$ is determined. The mass of the absorbing atom is called M_1 , and the scattering atomic mass is M_2 . For the calculation, we assume that the atomic mass is concentrated in the centre of the pair of absorbing and scattering atoms. The potential $V_{\text{eff}}^A(x)$ will take the form

$$V_{\text{eff}}^A(x) = V(x) + \sum_{j \neq i} V\left(\frac{\mu}{M_i} x \hat{\mathbf{R}}_{12} \cdot \hat{\mathbf{R}}_{ij}\right), \quad (9)$$

where $V(x)$ represents an interaction potential between absorber and backscatter atoms, the sum of i over absorber ($i = 1$) and backscatter ($i = 2$) atoms and the sum of j over their nearest neighbours describe the lattice contributions to pair interactions and depend on the crystal structure type. $\hat{\mathbf{R}}$ is the unit vector and $\mu = M_1 M_2 / (M_1 + M_2)$ is the reduced mass. For simplicity, we assume $M_1 = M_2 = M$ and $\mu = M/2$. For fcc crystals, we have

$$V_{\text{eff}}^A(x) = V(x) + 2V\left(-\frac{x}{2}\right) + 8V\left(-\frac{x}{4}\right) + 8V\left(\frac{x}{4}\right). \quad (10)$$

Similarly, the values of k_{eff}^D and $k_{3\text{eff}}^D$ in the ADM can be obtained by determining the $V_{\text{eff}}^D(x)$ potential with an expression of the single-particle effective potential, and when only the influence of N neighbour atoms is taken into account,

the $V_{\text{eff}}^{\text{D}}(x)$ potential can be written as

$$V_{\text{eff}}^{\text{D}}(x) = \sum_{j=1}^N V(x\hat{\mathbf{R}}^0 \cdot \hat{\mathbf{R}}_j), \quad (11)$$

where $\hat{\mathbf{R}}^0$ has been defined above, and $\hat{\mathbf{R}}_j$ is the unit vector of the j th atom from the equilibrium position. For single-atom fcc crystals,

$$V_{\text{eff}}^{\text{D}}(x) = V(x) + V(-x) + 4V\left(\frac{x}{2}\right) + 4V\left(-\frac{x}{2}\right). \quad (12)$$

The use of the effective harmonic potentials given in the above equations converts the complex three-dimensional problem for multi-particle effects into a simpler one-dimensional problem.

The Morse potential is expanded to the third-order around its minimum point:

$$V(x) = D(e^{-2\alpha x} - 2e^{-\alpha x}) \approx D(-1 + \alpha^2 x^2 - \alpha^3 x^3 + \dots), \quad (13)$$

where α describes the width of the potential, and D is the dissociation energy. For two-component intermetallic alloys AB, if atom A is represented by index 1 and atom B (blended) is represented by indication 2, then the width of potential will be α_{12} and the dissociation energy is D_{12} . Values of α_{12} and D_{12} are calculated as a percentage of alloy doping [8]. In equation (13), the other parameters have been defined as $y = x - a$, $x = r - r_0$, $a = \langle r - f_0 \rangle$, where, r is the distance between two atoms at temperature T , and r_0 is its value at the equilibrium position. The atomic vibration is quantized as phonon, that is why we express y in the term of annihilation and creation operators, \hat{a} and \hat{a}^+ , i.e., $y = a_0 = (\hat{a} + \hat{a}^+)$; $a_0^2 = \hbar\omega_D^0/2k_{\text{eff}}$, and use the harmonic oscillator state $|n\rangle$ as the eigenstate with the eigenvalue $E_n = n\hbar\omega_D^0$, ignoring the zero-point energy for convenience. Using the quantum statistical method, where we have used the statistical density matrix Z and the unperturbed canonical partition function ρ_0 , $Z = \text{Tr} \rho_0$, we determined the MSRD, MSD function.

According to the anharmonic correlated Debye model, we have

$$k_{\text{eff}}^{\text{A}} = 5D_{12}\alpha_{12}^2, \quad k_{3\text{eff}}^{\text{A}} = -\frac{3}{4}D_{12}\alpha_{12}^3, \quad (14)$$

and the anharmonic Debye model is

$$k_{\text{eff}}^{\text{D}} = 8D_{12}\alpha_{12}^2, \quad k_{3\text{eff}}^{\text{D}} = -D_{12}\alpha_{12}^3. \quad (15)$$

The considered quantities in the current theory are derived in expressions (4)–(6) based on the dualism of an elementary particle, i.e., its corpuscular and wave properties. Next, we describe the system in the Debye model involving all different frequencies up to the Debye frequency, each of which corresponds to a wave with a frequency $\omega(q)$ and wave number q that vary in the first Brillouin zone. Based on ACDM, $\sigma^2(T)$ has the form

$$\sigma^2(T) = \frac{\hbar c}{2\pi k_{\text{eff}}^{\text{A}}} \int_0^{\pi/c} \omega_{\text{A}}(q) \frac{1 + z_{\text{A}}(q)}{1 - z_{\text{A}}(q)} dq, \quad (16)$$

$$z_{\text{A}}(q) = e^{(\beta\hbar\omega_{\text{A}}(q))}, \quad \omega_{\text{A}}(q) = 2\sqrt{\frac{2k_{\text{eff}}^{\text{A}}}{M}} \left| \sin\left(\frac{qc}{2}\right) \right|, \\ \beta = \frac{1}{k_{\text{B}}T}. \quad (17)$$

If we substitute $k_{\text{eff}}^{\text{A}}$ from equation (4) into equations (16), (17), it has the following form $\sigma^2(T)$:

$$\sigma^2(T) = \frac{\hbar c}{10\pi D_{12}\alpha_{12}^2} \int_0^{\pi/c} \omega_{\text{A}}(q) \frac{1 + z_{\text{A}}(q)}{1 - z_{\text{A}}(q)} dq, \quad (18)$$

$$z(q) = e^{(\beta\hbar\omega_{\text{A}}(q))}, \\ \omega_{\text{A}}(q) = 2\sqrt{\frac{10D_{12}\alpha_{12}^2}{M}} \left| \sin\left(\frac{qc}{2}\right) \right|, \\ |q| \leq \frac{\pi}{c}. \quad (19)$$

Similarly, for ADM, the analytical expressions of $u^2(T)$ have been determined as

$$u^2(T) = \frac{\hbar c}{2\pi k_{\text{eff}}^{\text{D}}} \int_0^{\pi/c} \omega_{\text{D}}(q) \frac{1 + z_{\text{D}}(q)}{1 - z_{\text{D}}(q)} dq, \quad (20)$$

$$z_{\text{D}}(q) = e^{(\beta\hbar\omega_{\text{D}}(q))}, \\ \omega_{\text{D}}(q) = 2\sqrt{\frac{2k_{\text{eff}}^{\text{D}}}{M}} \left| \sin\left(\frac{qc}{2}\right) \right|, \quad (21)$$

If we substitute $k_{3\text{eff}}^{\text{D}}$ from equation (15) into equations (20), (21), we obtain

$$u^2(T) = \frac{\hbar c}{16\pi D_{12}\alpha_{12}^2} \int_0^{\pi/c} \omega_{\text{D}}(q) \frac{1 + z_{\text{D}}(q)}{1 - z_{\text{D}}(q)} dq, \quad (22)$$

$$z_{\text{D}}(q) = e^{(\beta\hbar\omega_{\text{D}}(q))}, \\ \omega_{\text{D}}(q) = 2\sqrt{\frac{8D_{12}\alpha_{12}^2}{M}} \left| \sin\left(\frac{qc}{2}\right) \right|, \\ |q| \leq \frac{\pi}{c}, \quad (23)$$

where c is the lattice constant, q is the phonon wave number, and M is the mass of composite atoms. From equations (18), (19), (22), (23), and (6), we have the correlated function $C_{\text{R}}(T)$, which has the following form for any crystal structure:

$$C_{\text{R}}(T) = \frac{\hbar c}{2\pi} \left\{ \frac{1}{k_{\text{eff}}^{\text{D}}} \int_0^{\pi/c} \omega_{\text{D}}(q) \frac{1 + z_{\text{D}}(q)}{1 - z_{\text{D}}(q)} dq \right. \\ \left. - \frac{1}{k_{\text{eff}}^{\text{A}}} \int_0^{\pi/c} \omega_{\text{A}}(q) \frac{1 + z_{\text{A}}(q)}{1 - z_{\text{A}}(q)} dq \right\}, \\ |q| \leq \frac{\pi}{c}. \quad (24)$$

If we substitute Morse potential parameters for fcc crystals into equation (24), we obtain the correlated function

Table 1. Morse potential parameters calculated by the current theory and experimental values.

Quantities/Crystals	$D_{12}(\text{eV})$ (Present)	$D_{12}(\text{eV})$ (Expt. [10])	$\alpha_{12}(\text{\AA}^{-1})$ (Present)	$\alpha_{12}(\text{\AA}^{-1})$ (Expt.[10])
Cu-Cu	0.3429	0.3528	1.3588	1.4072
CuAgX (X = 72)	0.3381	—	1.3634	—
CuAgX (X = 50)	0.3376	—	1.3638	—

Table 2. Effective force constants calculated by the current theory and experimental values.

Quantities/ Crystal	$k_{\text{eff}}^A(\text{eVA}^{-2})$ (Present)	$k_{\text{eff}}^A(\text{eVA}^{-2})$ (Present)	$k_{\text{eff}}^D(\text{eVA}^{-2})$ (Present)	$k_{\text{eff}}^D(\text{eVA}^{-2})$ (Expt.)	$k_{\text{3eff}}^A(\text{eVA}^{-3})$ (Present)	$k_{\text{3eff}}^A(\text{eVA}^{-3})$ (Expt.)	$k_{\text{3eff}}^D(\text{eVA}^{-3})$ (Present)	$k_{\text{3eff}}^D(\text{eVA}^{-3})$ (Expt.)
Cu-Cu	3.1655	3.4931	5.5889	5.7520	0.6646	0.8070	3.0889	0.9831
CuAgX (X = 72)	3.1423	—	5.0278	—	0.6814	—	2.6874	—
CuAgX (X = 50)	3.1396	—	5.0234	—	0.6423	—	0.8569	—

$$C_R(T) = \frac{\hbar c}{2\pi D_{12} \alpha_{12}^2} \left\{ \frac{1}{4} \int_0^{\pi/c} \omega_D(q) \frac{1 + z_D(q)}{1 - z_D(q)} dq - \frac{1}{5} \int_0^{\pi/c} \omega(q) \frac{1 + z_A(q)}{1 - z_A(q)} dq \right\},$$

$$|q| \leq \frac{\pi}{c}. \tag{25}$$

3. Numerical results and discussion

We applied equations (18)–(25) to the numerical calculation for Cu crystal and CuAgX (X = 72; 50) alloys. The results of the theoretical calculation of the Morse potential parameter and the experimental Morse parameters [10] are presented in table 1, and the elastic force constants are presented in table 2. A comparison of the data shows the agreement of the theoretical calculations with experimental values and other studies [9, 14, 10, 11]. By substituting the parameters in tables 1 and 2 into equations (18), (22), (25), we will get the mean square relative displacement $\sigma^2(T)$, the mean square displacement $u^2(T)$, and the correlation displacement function $C_R(T)$ of Cu crystals and CuAgX (X = 72; 50) alloys.

According to the data presented in tables 1 and 2, there is a significant difference between the correlation oscillation model and the single-particle anharmonic oscillation model. The force constant of single-particle anharmonic oscillation (ADM) k_{eff}^D is much greater than k_{eff}^A of the correlated oscillator (ACDM). The reason for this difference is the determination of the number and mass of atoms oscillating in two models. For the correlation oscillation model, the number and mass of atoms are only half those of the single-particle anharmonic oscillator. If the mass is considered to be concentrated in the centre of a mono couple bond for a correlated oscillation model, a crystal will act as quasi-atoms, which means that the mass is reduced to equal only half of the composite atomic mass, as shown in equation (11). The

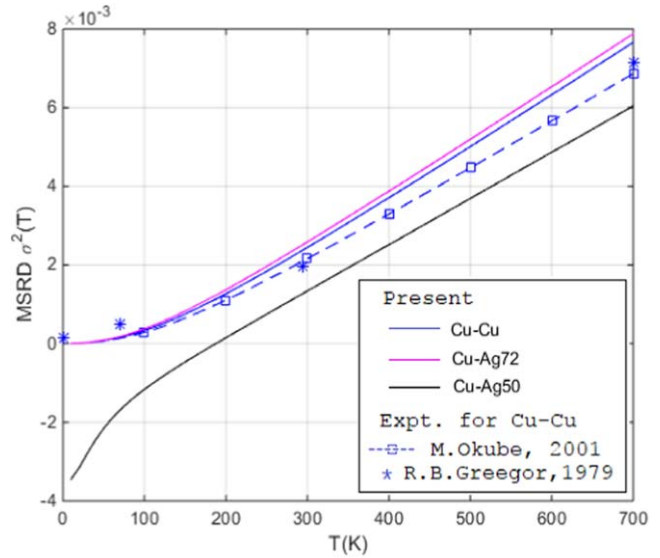


Figure 1. Dependence on temperature of mean square relative displacement $\sigma^{(2)}(T)$ for Cu, CuAgX (X = 72; 50).

number of atoms is only half the number of atoms for a single-particle anharmonic oscillation model because each quasi-atom is made up of a pair of composite atoms.

Figures 1 and 2 depict the temperature dependence of $\sigma^2(T)$ and $u^2(T)$ for Cu and CuAgX (X = 72; 50). They show a linear relationship to the temperature T at high temperatures. However, values of $\sigma^2(T)$ are greater than values of $u^2(T)$ at the same temperature, which is evident in figure 2. The experimental values of $\sigma^2(T)$ (points *) are higher with the experimental line of $u^2(T)$ [11].

Figure 3 illustrates the temperature dependence of the correlation function $C_R(T)$ of Cu crystal and alloys of CuAgX (X = 72; 50). Similar to the graphs depicting the temperature dependence of $\sigma^2(T)$ and $u^2(T)$, they are all linearly proportional to the temperature T at high temperatures, where the classical limit is applicable. At low temperatures, the curves for Cu and CuAgX (X = 72) contain zero-point energy contributions - a quantum effect. The calculated results of

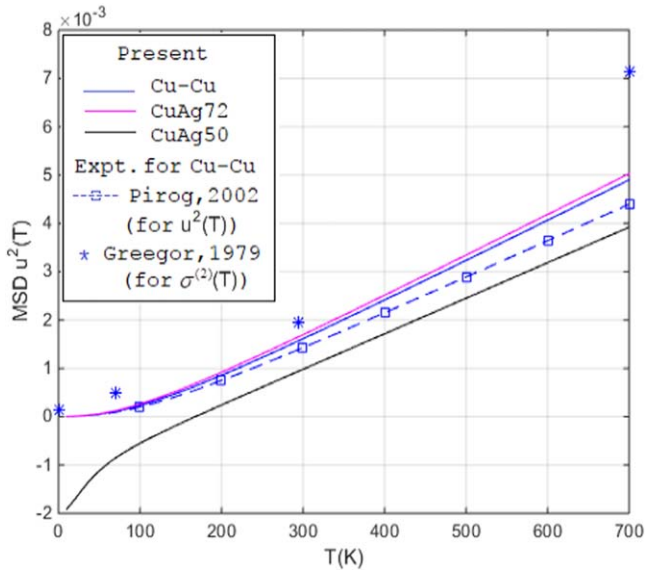


Figure 2. Dependence on temperature of mean square relative displacement $u^2(T)$ for Cu, CuAgX ($x = 72; 50$).

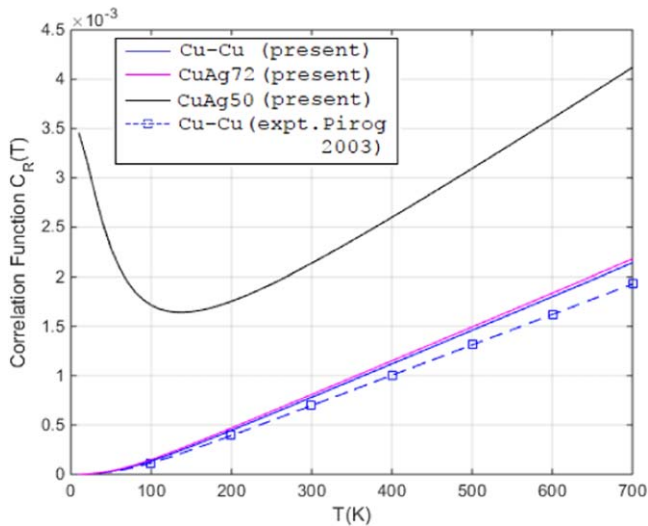


Figure 3. Temperature dependence of the correlation function $C_R(T)$ for Cu, CuAgX ($X = 72; 50$).

$\sigma^2(T)$, $u^2(T)$ for the Cu crystal fit well with the experimental values [10], [11]. Thus, it is possible to infer that the calculation results of the current theory for CuAgX ($X = 72; 50$) are reasonable. Moreover, the values of $\sigma^2(T)$ are greater than those of $u^2(T)$, making the extinction coefficients in EXAFS of the correlation oscillation model larger than those of single-particle anharmonic oscillation models.

An interesting feature in all three graphs is the curve of CuAgX ($X = 72; 50$) intermetallic alloys. For the CuAgX ($X = 72$) alloy, the graph curve format is similar to the curve of pure Cu crystal, meaning that the structure of the CuAgX ($X = 72$) alloy is not broken, and the structure type is still fcc. However, for the CuAgX ($X = 50$) alloy, the graph has an irregular shape, does not zero-point energy, and does not follow the rules like Cu and CuAgX ($X = 72$) at low temperatures. At high temperatures (about over 200 K), the curve

gradually returns to linear forms like Cu and CuAgX ($X = 72$) crystals. It is speculated that for the CuAgX alloy at a ratio of 50:50, the atoms are no longer closely linked to each other in the style of the fcc lattice at low temperatures (meaning that no CuAgX alloy material exists in this ratio, $X = 50$). As the temperature increases, the correlation between the atoms changes until the temperature reaches a certain value (over 200 K). The fcc lattice order slowly recovers, and the graph curve of CuAgX ($X = 50$) is linear to the temperature at high temperatures, similar to the results for Cu and CuAgX ($X = 72$). This is entirely consistent with studies done with other model theories and experiments with the CuAgX alloy at the ratio of $X = 50$ [8, 12].

4. Conclusions

In this work, a method was deduced to analyze the correlation effects of the $C_R(T)$ displacement correlation function based on DWF, the mean square relative displacement $\sigma^2(T)$ and the mean square displacement $u^2(T)$ in EXAFS spectra. The theory was applied to pure metals with an fcc structure and their alloys with different ratios.

The analytical expressions of $C_R(T)$, $\sigma^2(T)$, and $u^2(T)$ were inferred based on Debye models. The advantage of these models is based on the use of anharmonic effective potentials, which take the contributions of all of the nearest neighbouring atoms into account. The difference in the effective elastic force constant causes a difference in the thermodynamic properties of the crystals, and the thermodynamic properties are described by the correlated oscillation model and the single-particle correlation oscillation model. The difference in the number and mass of vibrating atoms in these models causes the difference in thermodynamic properties.

The correlation effects and the quantities $\sigma^2(T)$, $u^2(T)$, and $C_R(T)$ are dependent on the temperature. Their values are all linearly proportional to the temperature T at high temperatures, where the classical limit is applicable. At low temperatures, they contain zero-point energy contributions - a quantum effect.

The crystal lattice of the CuAgX ($X = 50$) alloy showed an abnormal disorder at low temperatures, where Cu and Ag atoms no longer had linked lattice structures according to the fcc structure. This result discovered in the studies from the experiment [12] and some theoretical studies following other models [7, 8]. These anomalies may bring many new interesting in-depth studies for researchers specializing in materials science.

The good agreement between the calculation results of the current study and the values obtained from experiments and calculations by other models proves the effectiveness of the present theory in EXAFS spectrum data analysis, especially for the study of correlation effects.

Conflicts of interest

The authors declare that they have no conflicts of interest.

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References

- [1] Frenkel A I and Rehr J J 1993 Thermal expansion and x-ray-absorption fine-structure cumulants *Phys. Rev. B* **48** 585
- [2] Crozier E D, Rehr J J and Ingalls R 1988 *X-ray Absorption: Principles, Applications, Techniques of EXAFS, SEXAFS and XANES* (New York: Wiley) p 373 Chap. 9
- [3] Hung N V and Rehr J J 1997 Phys., Anharmonic correlated Einstein-model Debye-Waller factors *Rev. B* **56** 43
- [4] Stern E A, Livins P and Zhang Z 1991 Thermal vibration and melting from a local perspective *Phys. Rev. B* **43** 8850
- [5] Schowalter M, Rosenauer A, Titantah J T and Lamoen D 2009 Computation and parametrization of the temperature dependence of Debye–Waller factors for group IV, III–V and II–VI semiconductors *Acta Cryst. A* **65** 5
- [6] Beni G and Platzman P M 1976 Temperature and polarization dependence of extended x-ray absorption fine-structure spectra *Phys. Rev. B* **14** 1514
- [7] Duc N B 2020 Influence of temperature and pressure on cumulants and thermodynamic parameters of intermetallic alloy based on anharmonic correlated Einstein model in EXAFS *Phys. Scr.* **95** 075706
- [8] Ba Duc N and Quang Tho V 2019 Dependence of cumulants and thermodynamic parameters on temperature and doping ratio in extended x-ray absorption fine structure spectra of cubic crystals *Physica. B* **55** 1–5
- [9] Pirog I V, Nedoseikina T I, Zarubin A I and Shuvaev A T 2002 Anharmonic pair potential study in face-centred-cubic structure metals *J. Phys. Condens. Matter* **14** 1825
- [10] Okube M and Yoshiasa A 2001 Anharmonic effective pair potentials of group VIII and Lb Fcc metals *J. Synchrotron Radiat.* **8** 937
- [11] Greegor R B and Lytle F W 1979 Extended x-ray absorption fine structure determination of thermal disorder in Cu: comparison of theory and experiment *Phys. Rev. B* **20** 4908
- [12] Kraut J C and Stern W B 2000 The density of gold-silver-copper alloys and its calculation from the chemical composition *J. Gold Bulletin* **33** 52–5
- [13] Van Hung N, Bao Trung N, Ba Duc N, Duy Son D and Sy Tien T 2014 Debye-Waller factor and correlation effects in XAFS of cubic crystals *Journal of Physical Science and Application* **4** 43–9
- [14] Tranquada J M and Ingalls R 1983 Extended x-ray absorption fine-structure study of anharmonicity in CuBr *Phys. Rev. B* **28** 3520
- [15] Yokoyama T, Sasukawa T and Ohta T 1989 Anharmonic interatomic potentials of metals and metal bromides determined by EXAFS *Jpn. J. Appl. Phys.* **28** 1905