Disorder-induced metal-insulator transition in cooled silver and copper nanoparticles: A statistical study

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1. Introduction

The growing interest for the understanding of the remarkable physical and chemical properties of low-dimensional systems such as nanoparticles, two-dimensional (2D) materials, nanotubes (wires), and their evolution under certain conditions arises in part from the quest for new commercially viable technologies [1,2]. In particular, metallic nanoparticles, such as copper and silver nanoparticles, have attracted considerable attention owing to their potential applications in catalytic, biological, electronic, optical, and information technological fields [3,4]. Thus, several studies have been mainly carried out to understand the size dependence of their properties originated by quantum confinement effects [5–10]. For instance, Darugar et al.[7] experimentally showed that the hot electron energy relaxation is faster for small copper nanoparticles because of the size-dependent electron-surface phonon coupling. It has also been proved, by employing several theoretical methods, that silver and copper nanoparticles undergo a structural transition from icosahedral to fcc local structure after increasing the nanoparticle size [9,5]. Regarding the electronic properties, it has been reported that nanoparticles with diameter greater than \( \sim 3.5 \) nm (for copper) and \( \sim 4.1 \) nm (for silver) present similar electronic behavior to their corresponding macroscopic counterpart [9,10]. However, it has been scarcely studied how different types of disorder (e.g., structural, chemical, magnetic, among others) will affect the electronic character of metallic nanoparticles, which is of crucial importance for the development of nanoelectronics.

In practice, there exist various inevitable types of disorder in nanoparticles, which may originate during the preparation of the sample (doping, deformation, and/or topological defects) [6,1]. As it is well-known, Anderson-type disorder (chemical disorder) can induce the localization of the electronic wavefunctions (Anderson localization) and, hence, lead to a disorder-induced metal-insulator transition (MIT, also called Anderson transition) [11–14]. Initially, this transition was only verified to happen for three-dimensional (3D) materials [15–19]. However, recently, it has been experimentally found that a defect density of more than \( \sim 1.2\% \) induces to Anderson localization in a single-layer graphene (2D material) yielding a metal-insulator transition [20]. This is an experimental proof of the influence of disorder on the electronic character of graphene layers, as was predicted theoretically [21–23]. Thereby, it remains an open questions if it is possible to find a MIT in other novel (not 3D) materials. From a theoretical point of view, there are several methods to verify the existence of the Anderson transition but the most used one is to determine the critical disorder, \( W_c \), in which this transition happens by using tools of the random matrix theory (RMT) combined with the statement (from scaling theory) that physical properties are size independent.
at the critical point [16,24,25]. In fact, it has been found that \( W_c \) for the 3D Anderson model [16] (simple cubic lattice) and 3D quasiperiodic lattice [19] are 16.5 ± 0.5 and 21.2 ± 0.6, respectively. For nanoparticles (considered as zero-dimensional materials), up to our knowledge, it has been only reported, by analyzing statistical properties of the energy spectrum, a crossing between metallic and insulating regime properties after increasing the chemical disorder strength (our previous works) [26,9].

The purpose of the present paper is to show the existence of the metal-insulator transition in silver and copper nanoparticles by using the method explained above. We focus on two different types of disorder: structural and chemical. Structural disorder is produced by cooling the nanoparticles at a fast rate avoiding the system to reach the minimal configuration energy [27–29]. Whereas, chemical disorder is introduced in the (slow cooled) nanoparticles by randomly modifying the on-site energy of the tight-binding Hamiltonian employed to describe their electronic properties [26,16,21]. This study has been done for silver and copper nanoparticles that contain atoms ranging from 561 to 5083 (these numbers belong to the icosahedral magic numbers sequence) [30]. The solidification process of the nanoparticles was carried out by MD simulations using a suitable and effective semi-empirical potential for each element: a tight-binding (TB) potential for silver [31] and a Johnson potential (based on the embedded atom method, EAM) for copper [32], which accurately reproduce the thermodynamic and structural properties of most transition and noble metals. We found out that only chemical disorder changes the electronic character of these nanoparticles, from metallic to insulating. Subsequently, the critical disorder strength \( W_c \) for silver and copper nanoparticles are determined, verifying the MIT in these nanomaterials. In the next section, we describe the theoretical model and, then, the results of the influence of disorder on the electronic properties of silver and copper nanoparticles are discussed.

2. Theoretical model

Nanoparticles were obtained by performing molecular dynamics (MD) simulations with different (but the most effective) semi-empirical potentials for each chemical element. For silver nanoparticles, we use the many-body potential developed by Cleri and Rosato [31] on the basis of the second-moment approximation to the tight-binding model (SMA-TB). This potential has been successfully used in several studies such as the atomic and electronic structure transformation of silver nanoparticles under rapid cooling situations [27], a comparative study of freezing of silver clusters and nanowires [33], the structural and dynamics properties of Cu–Au bimetallic clusters,[34] the thermodynamic stability of nanometer-sized Cu–Au systems [35], and the influence of chemical disorder on the electronic states of the Ag5083 nanoparticle [26]. Whereas, the atomic interactions in copper nanoparticles have been simulated by using the Johnson’s potential [32], which is based on the Embedded Atom Method (EAM), originally developed by Daw and Baskes. The accuracy of this potential has been verified, e.g., in the study of the determination of the threshold of nanoparticle behavior in nano-sized copper [9], the binding energy and atomic structure of copper nanoparticles [36] and nanowires [37], the structural evolution of liquid aluminum [28] and silver [29] during rapid solidification processes.

The MD simulations of the nanoparticles are carried out for a cubic box without periodic boundary conditions, so that the nanoparticle surface is free. The equations of motion are integrated over time using the predictor-corrector algorithm. The step time used is 2.5 fs. We have considered silver and copper nanoparticles of different sizes which contain atom numbers, \( N \), ranging from 561 to 5083. The sizes have been selected so that all belong to geometric magic numbers with icosahedral symmetry, this was realized in order to promote structural stability [5,30].

Before cooling, we have equilibrated the nanoparticles at their liquid state for 640 ps. After this, thirteen different configurations have been saved each 10.0 fs. Then, as it was mentioned before, to generate structurally disordered nanoparticles, we perform cooling processes at a fast cooling rate, \( k_b = 5.0 \times 10^{13} \) K/s. For comparison, we also generated nanoparticles with a high level of crystallization (structural order) by using a very slow cooling rate of \( k_b = 2.0 \times 10^{11} \) K/s. On cooling, the temperature was decreased to room temperature (300 K) in steps of \( \Delta T = 2.5 \) K.

To model the electronic properties of nanoparticles, we have used a tight-binding picture as proposed in previous works [26,27,9,10]. We consider that the tight-binding electronic Hamiltonian, in the atomic orbital basis \( \{ | \mathbf{R} \rangle \} \) (with an orbital “s” for each atomic site), has the following matrix elements

\[
H_{\mathbf{R},\mathbf{R}' } = \langle \mathbf{R} | \mathbf{H} | \mathbf{R}' \rangle = \begin{cases} \varepsilon_{\mathbf{R}} & \text{if } | \mathbf{R}' - \mathbf{R} | \leq r_c, \\ t & \text{if } | \mathbf{R}' - \mathbf{R} | > r_c. \end{cases}
\]

with \( \varepsilon_{\mathbf{R}} \) and \( t = 1.0 \) eV as the on-site energy and hopping parameter, respectively. We define \( r_c (Ag) = 3.16 \) Å and \( r_c (Cu) = 2.77 \) Å as the average distance to nearest neighbors (from a given atom). The atomic positions \( \{ | \mathbf{R} \rangle \} \) are known from the molecular dynamics simulations and fixed to determine the electronic properties. Then, the eigenvalues, \( \{ E_i \} \) \( \{ i = 1, 2, \ldots, N \} \), and eigenvectors, \( \{ \psi(E_i) \} \), are obtained from direct diagonalization of the Hamiltonian using LAPACK library.

Chemical disorder is introduced in the nanoparticles by modifying the on-site energy of the Hamiltonian (see Eq. (1)), which can be done by employing different disorder distribution [16,17]. In the present work, we focus on the box-like distribution, which consists in uniformly distributing \( \varepsilon_{\mathbf{R}} \) with random numbers \( g_{\mathbf{R}} \) within an energy window \( W \) (disorder strength), i.e.,

\[
\varepsilon_{\mathbf{R}} = \varepsilon_{\mathbf{R}}^0 + g_{\mathbf{R}} \in \left[ -\frac{W}{2}, \frac{W}{2} \right].
\]

The influence of chemical disorder in nanoparticles have been only studied for slow cooled nanoparticle in order to neglect the contribution from structural defects. The calculations shown in the present work (see Section 4) are the average over 30 random configurations for fixed disorder strength \( W \).

3. Spectral analysis

3.1. Participation number

To study the localization properties of an eigenvector \( \{ \psi(E_i) \} = \sum_{\mathbf{R}} c_{\mathbf{R}}^{\dagger} | \mathbf{R} \rangle \) of the Hamiltonian, we use the participation number

\[
P^{-1}(E_i) = \frac{1}{N} \sum_{\mathbf{r}} \left| c_{\mathbf{r}}^{\dagger} \right|^2,
\]

which measures the number of sites that significantly contribute to an electronic state of eigenvalue \( E_i \). The corresponding fraction of all the states is called the participation ratio, \( p \), which presents two limit cases: when \( p = 1 \) the wave function is fully extended, whereas for \( p = 1/N \), it is completely localized [38,19]. However, on averaging the results within a small energy interval, a more quantitative analysis of the participation number is possible. It is based on the power “q” of the size dependence of the participation
number, \( P > N^2 \). The exponent will be equal to 1 for extended states, and it will vanish for localized states. We average all the participation numbers in a small interval \( \Delta \epsilon = 0.2 \epsilon \) around the band center for all disorder strength \( W \) and perform a standard linear regression to fit the data of \( \log P \) versus \( \log N \) in order to determine \( q \) [22].

### 3.2. Level spacing statistics

Before calculating any statistical property of the energy spectrum, we must first apply an unfolding procedure in order to extract universal level statistics which can then be compared to the results of Random Matrix Theory (RMT) [16]. Following our previous works [26,9], we employ a sixth degree polynomial expansion to perform this procedure. Then, the energy level spacing is defined by \( s_1 = \epsilon_{im} - \epsilon_i (m = 1, 2, 3, 4) \), where \( \{ \epsilon_i \} \) are the so-called unfolded energies.

To analyze the influence of structural and chemical disorder on the electronic properties, we calculate the nearest-neighbor level spacing distribution (in units of the average level spacing \( \langle \delta \epsilon \rangle \)), \( P(s) \). This quantity has been used to study the electronic behavior in systems of current relevance, e.g., DNA [39], graphene phases [21–23], quasicrystals [38,40], and nanoparticles [26,9]. This is mainly because this distribution has two extreme behaviors with important physical meanings: for weakly disordered systems (metallic regime), \( P(s) \) is well described by the Wigner distribution, \( P_W(s) = \frac{1}{2} \pi \exp (-\frac{s^2}{2}) \), which is a good approximation to \( P(s) \) of the Gaussian Orthogonal Ensemble (GOE) from RMT. Whilst for strongly disordered systems (insulating regime), \( P(s) \) follows a Poisson distribution, \( P_P(s) = \exp (-s) \). Furthermore, for intermediate statistics, it has been developed several nearest-neighbor level spacing distributions to evaluate quantitatively the influence of disorder in a given system. The most used is the Brody distribution [41,42],

\[
P(s, \beta) = \alpha (\beta + 1) \pi^\beta \exp (-\pi^\beta s^{\beta+1}),
\]

with \( \alpha = \left( \frac{\Gamma (\frac{\beta+2}{\beta+1})}{\Gamma (\frac{\beta+1}{\beta+1})} \right) \beta+1 \) and \( \Gamma \) the well-known Gamma function. This distribution interpolates between the Poisson (\( \beta = 0 \)) and Wigner (\( \beta = 1 \)) ones. \( \beta \) parameter is related to the energetic repulsion among levels.

In order to have a better understanding of the electronic character of nanoparticles, we define the scaling parameter \( \alpha = J(s_1) \) [43], which is based on the integral of \( P(s, \beta) \), and \( J(s) \), reflects the strength of the repulsion of two consecutive levels when the separation between them is less than \( s_1 (s_1 = 0.473, \) first crossing point between \( P_W(s) \) and \( P_P(s) \) distributions), \( J(s) \) is expressed as,

\[
J(s) = \frac{I_W(s) - I_P(s)}{I_W(s) - I_P(s)}.
\]

where \( I_W(s) = \exp (-\pi s^2/4) \) and \( I_P(s) = \exp (-s) \) are the integral of \( P(s, \beta) \) corresponding to the two limiting regimes. This parameter also has two extreme cases, for a system with metallic character \( \alpha = \frac{W}{0.507} \), while for insulating systems \( \alpha = \frac{W}{0.569} \) [24]. Additionally, to study the influence of disorder on long order correlations among the electronic levels, we compute \( \Sigma \) statistics which measures the fluctuation in the number of energy levels \( n \) in an energy range \( L \) and it is defined as [16,40]

\[
\Sigma_2(L) = (\langle n^2(L) \rangle - \langle n(L) \rangle^2)^2.
\]

It is verified that for uncorrelated electronic levels (Poisson ensemble) \( \Sigma_2(L) = L \), whereas for a Gaussian Orthogonal ensemble with level repulsion \( \Sigma_2(L) \) increases logarithmically.

### 4. Results and discussion

#### 4.1. Structural disorder

Structural disorder can be detected by several ways. For instance, in Fig. 1(a), nanoparticles cooled at \( k_t = 5 \times 10^{13} \) K/s have a spherical shape with atoms randomly distributed on the surface, while nanoparticles cooled at slow rate \( k_t = 2 \times 10^{11} \) K/s show facets which are indicators of high level of crystallization in frozen materials [33,10]. Also, we have found that small slow cooled nanoparticles tend to show icosahedral external shape because it is the most stable geometry for the atom number that contains the system (icosahedral magic numbers) [9]. These facts are confirmed by the analysis of the radial distribution function, \( g(r) \) (see Fig. 1(c) and (d)). For Ag_{2057} and Cu_{2057} nanoparticles produced at \( k_t \), \( g(r) \) at 300 K shows the same shape as a liquid or amorphous material. Whereas, for slow cooled nanoparticles, we obtained a spiky distribution which is a fingerprint of crystallized (ordered) materials. Similar results were also reported for microstructures of silver [44,29] and copper [45] after solidification process at various cooling rates. The position of the peaks for nearest neighbors are in agreement with the corresponding ones of their fcc bulk structure, as it is expected [44]. By comparing \( g(r) \) at 700 K for both elements, we have also found that copper nanoparticles display greater melting point than silver ones.

To quantify local structural order in nanomaterials, we have used the common neighbors analysis (CNA) technique [46], in which four indices \( (i,j,k,m) \) are defined to identify the local environment of atoms. Here, we only analyze the index 1421 which corresponds to fcc-like local structure and its abundance is represented by \( A(\%) \). As it can be seen in Fig. 1(b), independently of the nanoparticle size, the index 1421 has more than 50 % of abundance for the set of silver and copper nanoparticles cooled at \( k_t \). Whereas, for fast cooled nanoparticles, the abundance of fcc structure is less than 20%, which indicates the existence of other local structures besides fcc-like one. Specially, for materials composed by silver and copper atoms it has been reported a competition between icosahedral and fcc-like local structures during solidification processes [29,27]. Besides, we obtained that \( A(\%) \) increases with the nanoparticle size, i.e., nanoparticles acquire the structure of their corresponding bulk phase (fcc structure) [10,9].

Because of their similar atomic structure and electronic configuration, the electronic properties of silver and copper nanoparticles display very close trends under the influence of structural disorder. In Fig. 2(a), we show that the electronic density of states \( \langle n(E) \rangle \) for structurally disordered Ag_{2057} nanoparticle has the same shape as such corresponding to its liquid phase, while for the slow cooled nanoparticle, it has a high density peak around the band edge.

\[ E/t \sim -0.4, \] which is characteristic for a fcc structure [9,47]. Moreover, it was found that, independently of the cooling rate, the level spacing distribution for these nanoparticles before and after cooling does not show any abrupt change and it has the shape of a Wigner-like distribution (see Fig. 2(b)). By employing other forming mechanisms, it has been also reported that small fcc-type particles display similar \( P(s) \) around the Fermi energy [48,47]. Unlike highly symmetric 1D and 2D systems [18], \( P(s) \) for the studied nanoparticles presents low probability of zero spacings, i.e., few degenerated levels. This is because the coexistence of many local structures which breaks the symmetry of the Hamiltonian. To quantify the influence of structural disorder on the electronic properties, we used the statistical parameters described in Sec. 3. We found that the influence of structural disorder on Brody and \( \alpha \) parameters is stronger for silver and copper nanoparticles with \( N < 2000 \) (see Fig. 2(c) and (d)) than for larger nanoparticles. However, the values remain in the range to be considered inside the
metallic regime (0.5 < \beta < 1.2) [42]. It can be also seen that structurally disordered nanoparticles display statistical parameters closer to those corresponding to the Wigner distribution (\beta \sim 0.94 and \xi \sim 0.508), i.e., they may conduct more current than the crystallized nanoparticles. It is worth mentioning that our results of the size dependence of statistical parameters differ from those found for the 2D Anderson model, continuous crossover from Wigner to Poisson distribution after increasing the system size [49]. Hence, structural disorder produced by modifying the cooling rate is not strong enough to induce a metal-insulator transition in the nanoparticles.

4.2. Chemical disorder

To study the influence of chemical disorder on the electronic properties of silver and copper nanoparticles, we have only considered nanoparticles obtained at \( k_S = 2 \times 10^{13} \) K/s. Thus, all the nanoparticles are structurally ordered and present an atomic structure very close to their corresponding macroscopic phase (fcc structure). In Fig. 3, we plotted the participation ratio, \( p(E) \), and electronic density of states, \( \eta(E) \), for \( Ag_{2057} \) nanoparticle at different disorder strength \( W/t \). The participation ratio at \( W/t = 0.0 \) indicates that the character of the electronic states is constant (\( p \sim 0.3 \)) but complex in almost the whole energy spectrum (see Fig. 3(a)). Moreover, despite having high density close to the band edge, the electronic states tend to be localized, i.e., \( p \sim 1/2057 \sim 0.0005 \). We have found that \( \eta(E) \) continuously decreases due to the spectral broadening and \( p(E) \) rapidly decreases to small values for all the states in the spectrum after increasing disorder. Based on the behavior of \( \eta(E) \) at the edge and central regions of the spectrum, it can be seen that correlations among electronic levels are changing when \( W/t \) increases. Similar effect was numerically found for weakly disordered graphene layers [21–23]. Fig. 3(c) displays the variation of scaling parameter “\( q \)” as a function of the disorder strength for both set of nanoparticles. This calculation has been done around the band center, \( E/t = 0.0 \). We obtained that nanoparticles present extended electronic states for \( W/t < 12.0 \), i.e., \( q \sim 1 \). Whereas, for higher strengths, we have that \( q \sim 0 \) which verifies the localization of electronic states around the band center. For states at the band edges, the localization effect is faster. It is worth noting that \( q \) is different to zero for strong disorder strengths (\( W/t > 40.0 \)) due to the finite size effect of the nanoparticles. A similar phenomena was reported by T. Rieth and M. Schreiber for a 3D quasiperiodic lattice (Amman-Kramer lattice) [19].

As it was discussed above, independently of the size, silver and copper nanoparticles display a level spacing distribution close to the Wigner distribution. Hence, to perform a better statistical analysis, from now on, we will focus on nanoparticles with \( N > 900 \) which have a Brody parameter of \( \alpha \sim 0.9 \). In Fig. 4(a), it can be seen that, when chemical disorder is added to copper nanoparticles, \( \beta \) presents three regions. In the first region (\( W/t < 12.0 \)), where nanoparticles are weakly disordered, \( P(\beta) \) remains close to \( P_{W}(\beta) \) (metallic character) and \( \beta \) values are very close to those expected in systems that belong to GOE (\( \beta = 0.95 \)) [41]. Moreover, in this region \( \beta \) for \( Cu_{561} \) and \( Cu_{2057} \) nanoparticles reaches its maximum value (\( W/t \sim 5, \beta = 0.94 \)). In the second region...
the chemical disorder which means that $P(s)$ for the nanoparticles crosses to a Poisson-like distribution (see inset of Fig. 4(a)). Finally, in the third region ($W/t > 40.0$), nanoparticles present an insulating character which is expected for strongly disordered systems [21]. This crossover, from metallic to insulating regime, has also been observed in graphene layers [21–23], 3D Anderson model [16], and other metallic nanoparticles [26,9]. Alike the work done by Hofstetter et al. [16], we have found that after increasing the nanoparticle size, $\beta(W)$-curve tends to be a step function; as is expected in the thermodynamic limit. However, despite the strong disorder in the nanoparticles, the Brody parameter cannot be zero because of the finite size effect [18]. Because of the similarities with the 3D Anderson model, we have employed the method introduced in Refs. [16,24,25] to determine the critical disorder, $W_C/t$, for the metal-insulator transition in the studied nanoparticles. This method is based on tools from the random matrix theory (RMT) and concepts of the scaling theory. In fact, we have found that the critical disorder, in which $\beta$ is size independent, is $W_C/t \sim 24.0$ and $W_C/t \sim 26.0$ for copper and silver nanoparticles, respectively (see Fig. 4(a) for the case of Cu NPs). These values are different from those found for a cubic lattice ($16.5 \pm 0.5$) [16,24,43] but close to the corresponding for a 3D quasiperiodic lattice ($21.2 \pm 0.6$) [19].

To strength the idea of a metal-insulator transition in nanoparticles, we have computed $\Sigma_p(L)$ statistics. We found that for silver and copper nanoparticles without disorder, $\Sigma_p(L)$ is well described by the Gaussian Orthogonal Ensemble (GOE) [16,40], Whereas, $\Sigma_p(L)$ crosses to the behavior expected for an insulating system (Poisson ensemble) after increasing the chemical disorder (see

Fig. 2. (a) Electronic density of states and (b) level spacing distribution for Ag$_{2057}$ nanoparticle at 300 K after cooling process at $k_s \sim 5 \times 10^{13}$ K/s (red line) and $k_F \sim 2 \times 10^{17}$ K/s (blue line). We also show the corresponding quantities for their liquid phase (dashed line). (c) Brody parameter and (d) scaling parameter $\alpha$ as a function of the atom numbers ($N$) for cooled nanoparticles. Expected value of $\alpha$ parameter for Wigner distribution is indicated by dashed lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 3. Variation of the (a) participation ratio and (b) electronic density of states for Ag$_{2057}$ nanoparticle after increasing the chemical disorder. (c) Scaling factor of the participation ratio as a function of the disorder strength for silver and copper nanoparticles.
This phenomena has also been found in the 3D Anderson model by studying the Wigner distribution from metallic to insulating regime. In the third region, as it also happened with the chemical disorder. The existence of metal-insulator transition in the nanoparticles for the engineering of highly efficient electronic nanodevices.

5. Conclusions

We have performed a statistical study to verify the existence of the metal-insulator transition in cooled silver and copper nanoparticles under the influence of structural and chemical disorders. It was found that structural disorder produced by modifying the cooling rate during solidification process is not strong enough to affect their electronic character, and it remains in the metallic regime. However, the electronic properties of nanoparticles crossed to the insulating regime after increasing the chemical disorder strength. Then, by applying the method introduced by Hofstetter et al. [16], we have found that the critical disorder $W_c/t = 24.0$, in which MIT happens for silver and copper nanoparticles are $24.0 \pm 1.1$ and $22.3 \pm 0.9$, respectively. These value are different from those reported for 3D Anderson model and 3D quasiperiodic lattice, which reflects a dependence on the local atomic structure on $W_c/t$. Moreover, we have verified the universality of the metal-insulator transition in the nanoparticles by finding this critical parameter for different disorder distributions. Hence, we expect our results to shed light on the applicability of metallic nanoparticles for the engineering of highly efficient electronic nanodevices.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.cplett.2017.05.048.

References