In situ pressure study of Rb$_4$C$_{60}$ insulator to metal transition by Compton scattering

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Compton scattering has been shown to be a powerful tool for studying the ground state electronic density in real materials. Using synchrotron radiation, we have studied pressure effects on Rb$_4$C$_{60}$ by measuring the Compton profiles below and above the insulator to metal transition at 0.8 GPa. The experimental results are compared with the corresponding calculated results, obtained from new ab initio energy band structure calculations. These results allow us to quantitatively evaluate contributions to the Compton profiles resulting from the contraction of the unit cell as well as from the contraction of the C$_{60}$ molecule itself. In this paper, we point out an unexpected contraction of the volume of the C$_{60}$ molecule, leading to a major effect on the electronic density of the Rb$_4$C$_{60}$ compound.

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I. INTRODUCTION

The family of compounds, A$_n$C$_{60}$ (A=K, Rb and Cs; n=1, 3, 4, and 6) exhibits a diversity of structural and electronic properties that has made them the subject of a great deal of interest. They exhibit conductivities ranging from metallic to insulating as a function of the alkali ion concentration.\(^1\) Recall that the neutral C$_{60}$ molecule possesses a closed shell electronic structure in its ground state. Therefore, within a rigid band picture, the threefold degenerate molecular orbital $t_{2u}$ of the pristine C$_{60}$ is partially occupied by electrons provided by the alkali atoms, resulting in metallic behaviors for A$_3$C$_{60}$ and A$_4$C$_{60}$ compounds, whereas the three suborbitals are filled for A$_6$C$_{60}$, leading to an insulator. In particular, the unusual behavior of the A$_5$C$_{60}$ family has intrigued the fullerene community. To begin with, x-ray powder patterns indicate that the contraction of the unit cell as well as from the contraction of the C$_{60}$ molecule itself. In this paper, we point out an unexpected contraction of the volume of the C$_{60}$ molecule, leading to a major effect on the electronic density of the Rb$_4$C$_{60}$ compound.

Furthermore, A$_4$C$_{60}$ compounds undergo an insulator to metal transition under pressure around 0.8 GPa, observed by the NMR study of Rb$_4$C$_{60}$,\(^9\) performed under pressure up to 12 kbar. Surprisingly, this observation did not lead to further investigation until our diffraction experiment, which exhibits an abrupt jump in the compressibility between 0.5 and 0.8 GPa. We attributed this jump to a structural phase transition preserving the tetragonal symmetry.\(^10\) In the present study, we go beyond the determination of the equation of states and focus on the study of the electronic density modification produced under pressure. Thus we have performed simultaneous Compton scattering and diffraction experiments, at pressures below and above the insulator-metal transition (i.e., 0.2 and 2 GPa). This enabled us to follow the electronic density change in momentum space caused by this phase transition. The wave functions obtained from ab initio energy band calculations of Rb$_4$C$_{60}$ were used to calculate Compton profiles be-
low and above the transition. Compton scattering is a bulk probe and, due to its incoherence, it is not sensitive to defects in the sample. Therefore it is particularly suitable for the investigation of synthetic materials. Furthermore, it is specially sensitive to delocalized states in the solid, i.e., valence and conduction electrons. We have already demonstrated the utility of this approach in the case of intercalated fullerenes. Since the information provided by Compton scattering about the ground-state electron distribution can be directly related to the Fourier expansion of the wave functions, it provides an excellent direct probe of the quality of calculated wave functions. Conversely, the calculated are used to understand and quantify the different electronic contributions to the Compton profile. These include the possible roles played by the $C_{60}$ molecular distortion, the contractions unit-cell volume and that of the $C_{60}$ molecular volume with pressure. The latter has been heretofore assumed to be negligible.

In Secs. II and III we present the Compton scattering method and the theoretical approach. In Sec. IV we describe the experimental procedures, including the sample preparation and characterization. The results are presented, discussed, and followed by concluding remarks in Sec. V.

II. COMPTON SCATTERING METHOD

Compton scattering involves the inelastic scattering of photons by electrons. The conservations of energy and momentum lead to a relationship between the energy loss of the photon and the projection of the initial electron momentum along the scattering vector $K$. The component of this energy loss resulting from scattering by the electrons at rest is referred to as the Compton shift. In addition, the motion of the electrons in the system results in a Doppler broadening of the distribution around the Compton shift and is a direct indicator of their initial momentum distribution.

In the impulse approximation (IA), that inelastic scattering process is assumed to be fast enough for the interaction potential to remain unchanged. Within this approximation, the Compton profile is defined as

$$J(q, e) = \int n(p) \delta(p \cdot e - q)dp = \int \chi^*(p)\chi(p)\delta(p \cdot e - q)dp,$$

where $e$ is the unit vector along the scattering vector $K$, $n(p)$ the electron momentum density, and $\chi(p)$ the electron wave function in momentum space, i.e., Fourier transform of the wave function in real space. Throughout the remainder of this paper we shall use atomic units (a.u.), for which $\hbar = m = 1$.

III. THEORETICAL APPROACH

The electronic structure of $Rb_4C_{60}$ was calculated first using the standard atomic positions, as determined by x-ray diffraction and, second, using those obtained by geometrical optimization. We employ the density functional theory in the local density approximation and using norm-conserving pseudopotentials. For carbon atoms, we use $2s$ and $2p$ as valence states. For rubidium atoms, we assumed an ionized configuration $4s^24p^64d^4$, treating $4s$, $4p$, and $4d$ as valence states. The wave functions were expanded in plane waves using a 60 Ry Cutoff. We used a $2 \times 2 \times 2$ Monkhorst-Pack grid for the electronic Brillouin zone (BZ) integration and a Gaussian smearing of 0.2 eV. The calculations were self-consistent, with no restriction on the form of charge density or potential. Due to the large size of the unit cell, four $k$ points were sufficient to achieve convergence in the BZ integration.

The calculated ground-state wave functions are represented by their plane wave expansion,

$$\psi_{n,k}(r) = \sum_{G} C_{n,k}(G) \exp[i(k + G) \cdot r],$$

where $G$'s are reciprocal lattice vectors. The large size of the primitive unit cell for this compound results in very short lengths of the reciprocal lattice vectors. Therefore the number of $G$'s necessary to obtain convergence in this sum is as large as 40,000. Using this expansion, Eq. (2) for the directional Compton profile takes the following form:

$$J(q, e) = \frac{1}{N} \sum_{n} \sum_{k} \sum_{G} |C_{n,k}(G)|^2 \delta((k + G) \cdot e - q) \partial(E_n - E_f).$$

The summation $G$ is over all the reciprocal lattice vectors for which the $C_{n,k}(G)$’s are non-negligible. The summation $k$ is over the symmetry-reduced sector of the BZ of the $Rb_4C_{60}$ compound is carried out using a tetrahedral interpolation method. The volume of this irreducible sector of the BZ is divided into tetrahedra by choosing a grid of $k$ points. The actual wave functions are calculated at each grid point and a linear interpolation is carried out for $|C_{n,k}(G)|^2$ within each tetrahedron. Due to the small size of the BZ for these compounds, a relatively coarse mesh of 13 points was sufficient for the BZ integration. The summation $n$ is over the occupied states. The function $\partial$ cuts off this summation at the Fermi energy in the case of a metal or a semi metal. Since the measurements are performed on powder samples, the comparison with experiment is made with an average theoretical profile obtained from four calculated directional profiles, i.e., (0 0 1), (0 1 0), (1 0 0), (1 1 a/c).

Our calculations were performed for a rigid configuration fixing the $C_{60}$ molecules in the most symmetric configuration, i.e., with the three double bonds chosen perpendicular to (100) directions and with the (111) directions passing through centers of hexagons. However, in a previous paper we reported the difference between Compton profiles of $C_{60}$ powder at temperatures below and above the orientational ordering phase transition (260 K). We showed that this difference was not larger than the statistical error. As a consequence, the average Compton profile resulting from the measurement on a powder sample is not sensitive to the orientational disorder of the $C_{60}$ molecule.
IV. EXPERIMENTAL PROCEDURE

We used a sample coming from the same origin as those used in the NMR measurement that exhibited the insulator-metal transition.\(^9\) The Rb\(_2\)C\(_{60}\) sample was prepared from a C\(_{60}\) powder. After rubidium doping up to saturation to form Rb\(_2\)C\(_{60}\), Rb\(_2\)C\(_{60}\) was obtained by reacting stoichiometric amounts of Rb\(_2\)C\(_{60}\) and C\(_{60}\), as described in Ref. 21.

The experiments were carried out using the high-energy beamline (Insertion Device 15B) of the European Synchrotron Radiation Facility in Grenoble, France.\(^2\) The synchrotron radiation beam was monochromatized to select 55.8 keV photons that were focused on a powder sample kept inside the pressure cell. This energy was chosen in order to optimize the positions of the Raman departures due to rubidium atoms with respect to the Compton peak position, i.e., to shift them as far as possible from the Compton peak. Rb\(_{1s}\) leads to a Raman departure at 9.4 a.u. (the low-energy side of the profile) and Rb\(_{2s}\) leads to a Raman departure at −11 a.u. (the high-energy side). We used a large volume apparatus (Paris-Edinburgh cell\(^2\)) as the pressure cell for combined diffraction and Compton measurements at pressures below and above the insulator-metal transition, i.e., at 0.2 GPa and 2 GPa, respectively.

The simultaneous diffraction measurements were performed for two different purposes: (i) to monitor cell parameters and sample symmetry; and (ii) to deduce the value of the pressure from the already known boron nitride equation of state (used as a pressure calibration standard).\(^2\) Diffraction patterns were collected in an angle-resolved geometry on an image-plate detector (MAR345). By moving the pressure cell perpendicular to the x-ray beam, we were able to obtain the spectra from the sample, the internal pressure calibrant (hexagonal boron nitride—hBN), and the sample environment (boron epoxy and hBN) successively. The two-dimensional diffraction images were analyzed using the ESRF Fit2D software, yielding intensity vs 2\(\theta\) patterns.

Compton scattering spectra were measured using a germanium detector under a scattering angle of 160 deg. In order to avoid all signals coming from the sample environment, a set of slits have been carefully designed, mounted before and after the sample, and checked in order to confine the scattering lozenge totally within the sample. The resolution function is deduced from the full width at half-maximum (FWHM) of the thermal diffuse scattering (TDS) peak and is equal to 0.46 a.u. After subtracting the background and correcting profiles from the energy dependent contributions (absorption in air, sample, and anvil cell, as well as detector efficiency), the wavelength scale was converted into momentum scale. Multiple elastic and inelastic scattering contributions (MSC) for each measured profile have been calculated using a Monte Carlo simulation, taking into account beam polarization, sample geometry, and density.\(^2\) MSC was then subtracted from measured profiles. Both measured spectra were normalized to the compound’s full number of electrons per carbon atom between −9.5 and 9.5 a.u., i.e., 8.13 electrons. Since many of the systematic errors cancel when one takes the difference between low- and high-pressure Compton measurements, the results are presented in the form of the difference between the two profiles.

V. RESULTS AND DISCUSSION

A. Experimental results

In Fig. 1, we show the measured Compton profile difference (CP difference), defined as the Compton profile measured at low pressure minus the Compton profile measured at high pressure. Five points are shown in the resolution range of 0.46 a.u., Here 9 \(\times\) 10\(^5\) and 2 \(\times\) 10\(^5\) counts have been integrated in a of 0.1 a.u. range at the top of the Compton profile for the low- and the high-pressure spectra respectively. Statistical error bars were calculated within a range of 0.1 a.u. The measured CP difference is symmetric up to 9.5 a.u., within the statistical accuracy of the experiment. As a consequence, the CP difference is symmetrized, leading to a reduction of the statistical error bars by a factor of 2.

Two main features on the CP difference should be pointed out. The maximum magnitude occurs at \(p_z\) = 0 a.u. and reaches 1% of the total profile, and the minimum is located around \(p_z\) = 1.3 a.u.

The following discussion will concentrate on the description of these two features by comparing them with the \textit{ab initio} calculations. The figure also gives the total profile at 0.2 GPa.

The high- and low-pressure cell parameters needed for the calculations are obtained from a detailed analysis of our x-ray diffraction 2\(\theta\) patterns. Our low-pressure cell parameters are in perfect agreement with previous measurements,\(^2\) i.e., \(a = 11.96 \text{ Å}\) and \(c = 11.02 \text{ Å}\). At high pressure we obtain \(a = 11.82 \text{ Å}\) and \(c = 10.63 \text{ Å}\),\(^10\) recalling that there is no structural change of symmetry under pressure.

We can rule out the possible polymerization, i.e., the breaking of double bonds through the formation of rings joining two C\(_{60}\) molecules, for the following two reasons. First, the diffraction patterns obtained\(^10\) do not show any evidence of polymerization between C\(_{60}\) molecules up to 5 GPa in contrast to that reported for C\(_{60}\).\(^{26}\) Second, if we compare the distance between the adjacent C\(_{60}\) molecules in Rb\(_2\)C\(_{60}\) and the polymerized compounds such as Li\(_4\)C\(_{60}\) or Na\(_4\)C\(_{60}\), we can conclude that Rb\(_2\)C\(_{60}\) is not polymerized in the pressure range used in this study. A simple geometrical
consideration provides C$_{60}$ molecule center-to-center distances in Rb$_4$C$_{60}$ equal to 10.09 Å at ambient pressure and 9.90 Å at 2 GPa. In contrast, the corresponding values are 9.13 Å and 9.28 Å for Li$_4$C$_{60}$ (double bonding monomers$^{27}$) and Na$_4$C$_{60}$ (singly bonded monomers$^{29}$), respectively. As a consequence, if we consider a diameter of C$_{60}$ molecule close to 7.1 Å, the C–C bond length between two adjacent molecules becomes equal to 2.80 Å for Rb$_4$C$_{60}$ at high pressure (2 GPa), 2.18 Å for Na$_4$C$_{60}$ and 2.03 Å for Li$_4$C$_{60}$. Third, C–C bond length values between 1.7 to 1.9 Å$^{29,30}$ are observed in other polymers formed by 2+2 cycloaddition such as RBC$_{60}$. Such values, which are also the highest possible values predicted by DFT in polymeric fullerides, are much lower than for Rb$_4$C$_{60}$. As a consequence, we infer that the C–C bond length in Rb$_4$C$_{60}$ is too large to indicate the cycloaddition process occurs, at least up to 2 GPa. Furthermore, if the C$_{60}$ molecule contracts under pressure, the C–C bond length between two adjacent molecules would become larger than 2.80 Å, making the polymerization even more unlikely.

B. Discussion

Three different components may contribute to the Compton profile difference.

(i) The so-called Jahn-Teller distortion, i.e., distortion of the C$_{60}$ molecule at ambient pressure. This distortion is assumed to be modified under pressure. It should be noted that this phenomenon has never been experimentally clearly observed, even if its role in driving A$_4$C$_{60}$ into an insulating phase is widely accepted.

(ii) The unit cell contraction under pressure, i.e., a decrease of cell-parameter values as measured by our diffraction experiments.

(iii) The volume contraction of the C$_{60}$ molecule under pressure.

In order to better understand the difference profile between low- and high-pressure measurements, we will present the effect of each of the three contributions listed above obtained by means of our LDA calculations.

1. Distortion of the C$_{60}$ molecule

a. Isolated C$_{60}$ molecule. We study the effect of the distortion of the C$_{60}$ “ball” resulting from the transfer of electrons from alkali atoms to the C$_{60}$ molecule. We define the diameter of the C$_{60}$ ball as the distance separating two carbon atoms symmetrically set on opposite sides of the molecule center. In the three cases below, we used the tight binding method$^{12}$ to generate relaxed positions of carbon atoms in an isolated C$_{60}$ molecule for different models of charge transfer.

(i) Neutral C$_{60}$ molecule. The t$_{1u}$ orbital is empty and the C$_{60}$ molecule remains spherically symmetric after relaxation. This case is hereafter referred to as the “neutral molecule” or “NM.”

(ii) C$_{60}$$^{4-}$ ion with nonuniform charge distribution. The first two levels of the unoccupied t$_{1u}$ are filled by the four electrons. Therefore occupation numbers of the two lower levels are 2 whereas it is 0 for the upper level. This removal of degeneracy leads to the Jahn-Teller distortion of the C$_{60}$ hereafter referred to as “JT.”

(iii) C$_{60}$$^{4-}$ ion with uniform charge distribution. Each of the three suborbitals of t$_{1u}$ is equally filled by the four electrons. Therefore the occupation number of each sublevel is 2/3. As a consequence, there is no distortion of the C$_{60}$ molecule, but only lengthening of all the C–C bonds accompanied by a uniform increase of its volume compared to neutral C$_{60}$. This case is referred to as the “uniformly charged molecule,” or “UCM,” in the following.

Figure 2 shows the values for the diameter of the C$_{60}$ molecule, as calculated for the three above cases. The result obtained in the UCM case, i.e., 7.14 Å, is in perfect agreement with the previously calculated results obtained by Erwin and Bruder.$^{33}$ The molecular distortion in the JT case leads to a dispersion of diameters between 7.13 and 7.16 Å. However, the average value of the JT diameters is very close to that of the UCM.

In order to determine the role played by the distortion of the isolated C$_{60}$ molecule in determining the features of the CP difference, we calculate Compton profiles for both JT and UCM cases. The carbon positions used were obtained from the JT and UCM cases, leading to calculated profiles J$_{JT}(p_x)$ and J$_{UCM}(p_x)$, respectively. In both cases we use rubidium atomic positions$^{2}$ used by Erwin and Bruder,$^{33}$ which we will refer to as standard positions in the following discussions.

Thus, the Compton profiles difference J$_{JT}(p_x)$−J$_{UCM}(p_x)$, shown in Fig. 3, indicates the Jahn-Teller distortion. Its magnitude is negligible [0.01 % of J(0)] compared to the experimental CP difference [1 % of J(0)]. In addition, one can notice that its features are out of phase with the experimental results (Fig. 1). In fact, our calculations have shown that Jahn-Teller distortion affects only individual diameters within C$_{60}$ molecule but not molecular volume itself. There-
Therefore, we can conclude that such a deformation, with a constant C$_{60}$ molecular volume, has a negligible effect on the CP difference.

In addition, in order to show the charge transfer effect on C$_{60}$ molecules, a second CP difference is shown in Fig. 3, i.e., $J_{\text{UCM}}(p_c) - J_{\text{NM}}(p_c)$, corresponding to the volume effect in an isotropic C$_{60}$ molecule geometry. $J_{\text{NM}}(p_c)$ was also constructed using standard rubidium atomic positions. The electronic charge transfer results in a rather small change of the volume of C$_{60}$ molecule, 0.42%, defined as $[(V_{\text{UCM}} - V_{\text{SM}})/V_{\text{UCM}}] \times 100$. Its contribution to the shape of the CP difference results in a minimum at 1.3 a.u., and by the normalization rule of CP, in a maximum at 0 a.u. Positions of these features are consistent with the experimental CP difference shown in Fig. 1.

Therefore, we conclude that Jahn-Teller distortion is not measurable by the Compton scattering method, but the Compton scattering profile is sensitive to an even smaller change in C$_{60}$ molecular volume.

b. Embedded C$_{60}$ molecule. The C$_{60}$ molecule (hereafter charged C$_{60}^{4-}$) is placed within the solid Rb$_4$C$_{60}$ environment, both at low and high pressures. Geometrical optimization was carried out within pseudopotential-LDA formalism to find new positions of carbon and rubidium atoms. Unit-cell parameters were chosen from our diffraction measurements at low as well as at high pressure. These two cases are referred to as “geometrically optimized at low pressure,” i.e., “GO(LP)” and “geometrically optimized at high pressure,” i.e., “GO(HP),” respectively. Figure 2 shows the values of the diameter for the embedded C$_{60}$ molecule. They are lower than those corresponding to the isolated molecule, and show a dispersion of 0.015 Å at low pressure and 0.02 Å at high pressure. Looking at low-pressure results, we notice that our calculations show that the C$_{60}$ ball is two times less distorted in the case of solid Rb$_4$C$_{60}$ (GO) than in the isolated case (JT).

In Fig. 4 we show the effect of pressure on the shape of Compton profile difference between low and high pressure, using geometrical optimized LDA calculations. We compare the calculated CP difference $J_{\text{GO(LP)}} - J_{\text{GO(HP)}}$ with the corresponding experimental result. We observe that the theoretical results do not match the experiment either in amplitude or in position of the measured minimum at $p_c = 1.3$ a.u.

Figure 2 shows the calculated values for C$_{60}$ diameters for both the isolated molecule as well as for the molecule embedded in the solid. We observe that the average values of the diameters for the GO molecules (for low as well as high pressure) are smaller by about 0.06 Å than those obtained for the isolated molecule. Kuntscher et al. showed by x-ray diffraction that the difference between the C$_{60}$ molecule equatorial radius and the polar radius (along the c axis) is less than 0.08 Å. The average value of 7.12 Å measured by them is close to the value of 7.10 Å obtained by Fleming et al. with an experimental accuracy to within 0.1 Å. We can therefore conclude that our calculated diameters as well as calculated dispersion values are compatible with experimental results, regardless of the surroundings, i.e., for an isolated C$_{60}$ molecule (neutral or charged) as well as C$_{60}$ surrounded by its solid environment.

2. Cell contraction under pressure

In this section, we follow the shape of the Compton profile as the Rb$_4$C$_{60}$ unit-cell volume decreases with pressure. Our aim is to evaluate the role of the cell-parameter contraction alone. For this purpose, we assume the C$_{60}$ molecule to be incompressible. Thus, we use the 60 carbon atom positions of the C$_{60}$ molecule as given for UCM in both low- and high-pressure calculations. The relative positions of ru-
The question is, how does this decrease of the molecular volume modify the shape of the Compton profile?

For this purpose, we take the difference between two calculated Compton profiles. First, using the 60 carbon atomic positions given in UCM with the standard Rb atomic positions \( p_{z} = 0 \) a.u., the amplitude of the experimental CP difference is higher than the calculated values by a factor 2.8. In addition, the position of the minimum for the experimental CP difference is located at 1.3 a.u., in contrast to 0.85 a.u. for the calculated result.

Going further, we attempted to find whether it was possible to obtain a better agreement between theory and experiment simply by using fictitious cell parameters corresponding to a higher pressure. Fitting the theoretical and the experimental CP difference at \( p_{z} = 0 \) a.u. corresponds to an experimental pressure as high as 9 GPa (i.e., \( a = 11.55 \) Å and \( c = 10.30 \) Å (Ref. 10)). The calculated difference, using these parameters, is shown in Fig. 5. We can see that the agreement between theory and experiment in the region around \( p_{z} = 1.3 \) a.u. is not significantly improved.

3. \( C_{60} \) molecular volume in the solid

In this section, we demonstrate the importance of \( C_{60} \) molecular volume variation for the shape of the calculated CP difference. We focus on what will happen to CP, hypothetically, if we keep both the pressure and \( Rb_{4} C_{60} \) unit-cell volume fixed, allowing only the change of the \( C_{60} \) molecular volume. In the following, we chose the fixed pressure to correspond to the low-pressure value.

Therefore, taking as a reference profile \( UCM(LP) \) calculations,\(^{33,36}\) we used a lower \( C_{60} \) molecular volume for the new \( UCM(LP) \) calculation than that corresponding to the reference profile. As an example, we chose the \( GO(LP) \) calculated diameter (7.14 Å) for the lower volume, a decrease of 0.06 Å (see Fig. 2). This value remains clearly within diffraction experimental accuracy, i.e., 0.1 Å (around 7.10 Å).

The question is, how does this decrease of the molecular volume modify the shape of the Compton profile?

C. Final results and concluding remarks

We notice that none of the UCM or GO models are able to describe the experimental CP change under pressure and that
This volume variation is unexpected since the C\textsubscript{60} molecule is usually considered as uncompressible. Nevertheless, our addition of these two contributions results in the difference \(J_{\text{GO}} - J_{\text{UCM}}\) at high pressure. We obtain a very good agreement: both the difference to the experimental CP difference between low and high pressure cannot be explained by taking into account a larger decrease of the C\textsubscript{60} molecule volume than given by any of these two models.

In conclusion, the shape of the Compton profile difference given by our measurements comes from both of the following:

(i) the contraction of the unit-cell due to pressure, contributing around 35\% to the magnitude of the PC difference at \(p_c=0\) a.u.

(ii) the contraction of the C\textsubscript{60} molecule under pressure that plays a role both on the magnitude (65\%) and position of the minimum.

The magnitude of the experimental Compton profile difference between low and high pressure cannot be explained without taking into account a contraction of the C\textsubscript{60} molecular volume under pressure. We have shown that a diameter contraction as large as 0.07 Å was required to reproduce the Compton profile difference. The high sensitivity of Compton scattering to the chemical bonding allows the detection of tiny variations of electronic momentum density related to contractions of the C\textsubscript{60} molecule diameter. This contraction cannot be seen by the present x-ray diffractions. In a previous paper, we demonstrated, through x-ray diffraction measurements as well as LDA calculations, that the insulator to metal transition is accompanied by an isostructural transition that also leads to an abrupt overall centering of rubidium atoms above C\textsubscript{60} molecule pentagons and hexagons. The Compton scattering experiment presented here provides the additional and significant indication of a C\textsubscript{60} volume contraction in the pressure range corresponding to an electronic transition. This last conclusion is based on the sensitivity of the Compton scattering to the outer electrons, i.e., electrons responsible for the bonding and subject to charge transfer. The significant change of C\textsubscript{60} volume that seems to occur reflects a change in electronic density, resulting in stronger intramolecular bonds than usually predicted.

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