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Experimental determination of the Compton profile of C_{60} through binary encounter electron spectroscopy

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The method of 0° electron spectroscopy was used to study binary encounter electrons resulting from hard collisions between 1.5 MeV/u C^{6+} ions and the electrons in a C_{60} vapor target. The Compton profile of C_{60} was then extracted from the electron spectra using an impulse approximation treatment. The experimental results are in excellent agreement with theoretical Compton profiles of C_{60}. The C_{60} Compton profile is compared with that of atomic carbon, as well as those for graphite and diamond. © *1995 American Institute of Physics.*

The past few years have seen a prodigious amount of research on fullerenes, C_{60} in particular. In general, the understanding of the many fascinating properties of fullerenes requires detailed knowledge about the electronic structure. First-principles calculations for C_{60} are in good agreement with photoemission and inverse-photoemission measurements.[1–3] On the other hand, simpler models[4–6] for the electronic structures contain in many cases the essential physical features to explain the experimental findings. The purpose of the present work is to present the experimental Compton profile of C_{60} and to use this Compton profile as a means of checking the accuracy of calculated C_{60} wave functions, in particular their momentum content.

One of the most direct experimental verifications of wave functions is the measurement of the Compton profile,[7–9] which is particularly sensitive to the details of the momentum distribution of the valence electrons. In the past, Compton profiles have also been shown[8] to be useful as

- a direct measurement of electron momentum distribution, projected onto the projectile axis (in this case averaged over all orientations);
- a measurement of the spatial electron density distribution (via Fourier transform), projected onto the projectile axis (in this case, averaged over all orientations);
- an indirect measurement of the total kinetic energy and, through the virial theorem, a measure of the total electronic energy of the system.

While the focus of this work is to provide a check on some published wave functions, the experimental results presented here may also be used to investigate other aspects of C_{60}, as mentioned above.

Normally, the Compton effect, that is, the inelastic scattering of photons, or ''e 2e,'' the angularly resolved inelastic scattering of electrons, is used to measure the Compton profiles of materials of interest. In this work, experimental Compton profiles of C_{60} were determined using the novel approach of studying the production of binary encounter electrons produced in high energy ion-C_{60} collisions. These Compton profiles were then compared with those generated from effective one-electron wave functions[4] for C_{60} as well as with the Compton profiles of graphite, diamond and atomic carbon.

The main emphasis of this work is the experimental C_{60} Compton profile per se and how it compares with the theoretical one. However, because the measurement technique is a novelty in itself, much of the discussion here is concerned with the experimental details, and the validity of the experimental approach.

The binary encounter electron (BEe) peak is a prominent feature[10] in the electron spectra of ion-atom and ion-molecule collisions. A qualitative model for BEe production was given several years ago.[11,12] More recently, a quantitative model[13,14] for BEe production based on the impulse approximation,[13] in which the target electrons (from C_{60} in this case) undergo elastic scattering from the projectile ion was given. In the case of a bare projectile ion, the elastic scattering is Rutherford scattering. In our discussions here we limit ourselves to 180° scattering (in the projectile frame, which corresponds to 0° in the lab frame) of the target electrons. For bare projectiles the doubly differential cross section for BEe production is given[15] by:

$$\left(\frac{d^2\sigma}{d\Omega\, dE}\right)^{\text{proj}}_{\text{BEe}} = \frac{J(Q)}{V_p + Q/m_e}\left(\frac{d\sigma}{dE}\right)^{\text{proj}}_{\text{Rutherford}}, \tag{1}$$

where $(d\sigma/dE)^{\text{proj}}_{\text{Rutherford}}$ is the Rutherford scattering cross section in the rest frame of the projectile, and V_p is the lab frame projectile velocity. The argument of the Compton profile, J, is Q, the component of the target electron's momentum projected along the collision axis. It is given by

$$Q = \sqrt{2m_e(E_{\text{proj}} + E_i)} - m_e V_p, \tag{2}$$

where E_i is the ionization potential of the target electron, and E_{proj} is the outgoing electron's energy measured in the projectile frame.

This simple model is valid so long as the velocity of the projectile is high enough that no rearrangement of the target electrons takes place during the collision (the impulse approximation). The model has been experimentally[14–16] found

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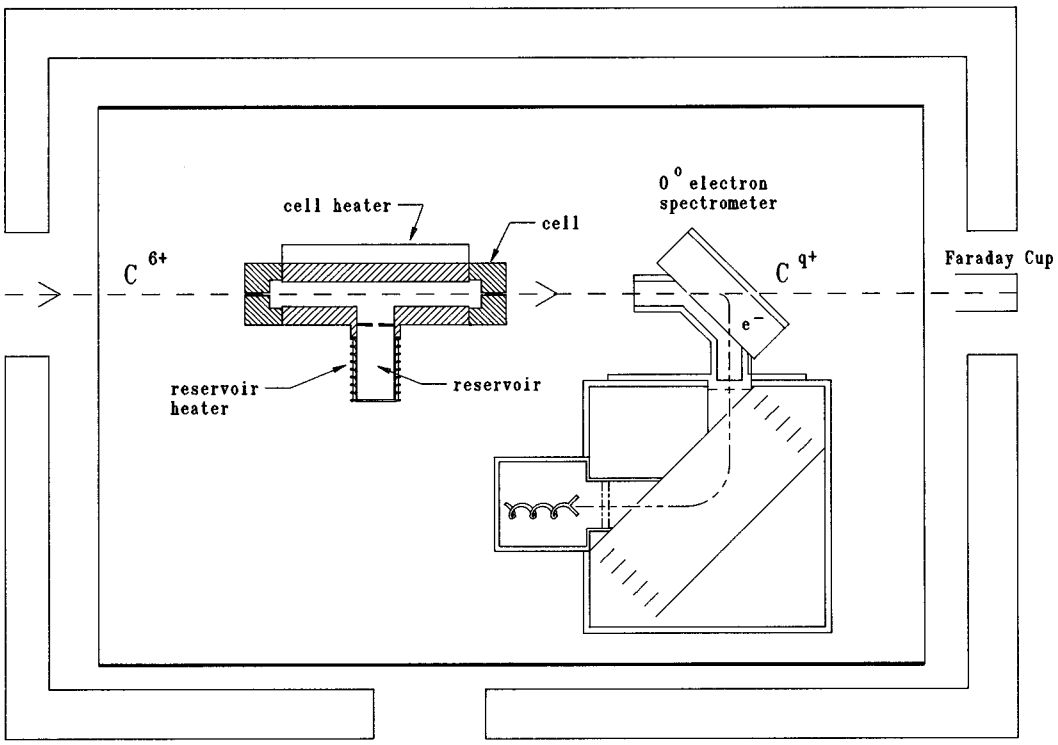

FIG. 1. Simplified overview of the experimental apparatus.

to work extremely well over a wide variety of projectiles and targets, and over a wide range of projectile energies.[17–19]

In eq. (2), a value of $7.48eV$, the first ionization energy of C_{60} is used for E_i. In a system as complicated as C_{60}, a whole range of ionization energies should be folded into eq. (2). Here, however, because of the high projectile velocity of 7.7 atomic units, corresponding to a projectile frame electron energy [E_{proj} in eq. (2) of nearly 830 eV], an uncertainty in the binding energy of a few tens of eV has a negligible effect on the *shape* of the measured Compton profile. Thus, even without a detailed knowledge of the C_{60} ionization energies, the technique of binary encounter electron spectroscopy should still produce a valid Compton profile. The major effect of using an incorrect effective ionization energy is having the peak of the extracted Compton profile shifted away from $Q=0$.

Fig. 1 shows a schematic representation of the experimental apparatus. It is essentially the same as used previously[20,21] in experiments with lithium vapor targets. Briefly, a collimated beam of 1.5 MeV/u C^{6+}, produced in the J.R. Macdonald Laboratory tandem Van de Graaff accelerator is directed through a C_{60} vapor-producing oven, and then through the first stage of a tandem 0° electron spectrometer, after which it is collected in a Faraday cup.

The tandem parallel plate electron spectrometer, which has an energy resolution, $\Delta E/E=2.5\%$ has been described previously.[22] The voltages on the spectrometer are computer controlled and the spectrometer dwells on each energy until a preset amount of charge has accumulated in the Faraday cup. The spectrometer can be operated in high resolution mode by decelerating the electrons in the region following the first 90° bend. For this work, high resolution mode was not used since the binary encounter peak is rather broad and structureless. In the lab frame energy range in which the binary encounter peak appeared, the spectrometer response function may be approximated by a Gaussian shape having a full width at half maximum of about 80 eV. Energy analyzed electrons are detected in a channeltron electron multiplier, the efficiency of which is flat over the energy range of interest.

The oven is a resistively heated stainless steel tube attached to a resistively heated reservoir. These were heated to 524°C and 513°C, respectively. This corresponds to a C_{60} partial pressure of roughly 1 mTorr.[23] Care was taken to insure oven temperatures had stabilized before data were taken. The fullerene powder placed in the oven reservoir was manufactured commercially,[24] and consisted of 90% C_{60} and 10% C_{70}.

Fig. 2 shows three typical spectra. Spectrum A is a ''background'' spectrum, taken with the oven heated to 410°C (corresponding to a C_{60} pressure of 0.02 mTorr). Spectrum B is taken with a nominal fullerene pressure of 1.0 mTorr. Spectrum C is spectrum A subtracted from spectrum B.

Several features are evident in spectra B and C. First of all, at about 270 eV one can see a small sharp structure. High resolution scans of this region reveal this to be a series of carbon K-Auger lines, resulting from target K-shell ionization by the projectiles. Similarly, at about 2100 eV are a series of projectile K-Auger lines, resulting from double electron capture by the projectiles, probably from the K-shell

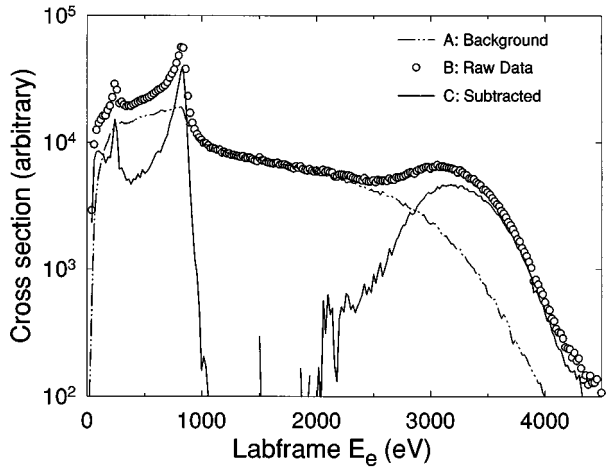

FIG. 2. Typical electron spectra. Spectrum A is a ''background'' spectrum. Spectrum B was taken with a nominal fullerene pressure of 1 mTorr. Spectrum C is spectrum A subtracted from spectrum B.

of the target carbon atoms. Though not evident in the low resolution spectra of Fig. 2, these projectile Auger lines become very clear when examined in high resolution. Projectile and target Auger spectra for this collision system will be the topic of a separate publication.

At about 830 eV is seen a large, sharp structure. This is known as the ''cusp,'' and represents electrons which have been captured to the continuum[25] of the projectile. Thus, these electrons move with the same velocity as the projectile, and therefore give a direct, very accurate measurement, of the projectile energy.

Finally, the broad ''hump'' centered at about 3300 eV is the binary encounter electron peak, the focus of this work. By inverting eq. (1) to solve for $J(Q)$, one obtains the experimental Compton profile plotted in Fig. 3. One may note at this point that because the background in Fig. 2 is higher at the low energy side of the binary encounter peak than at the high energy side, the corresponding uncertainty, both relative and absolute, in the subtracted spectrum is greater on the low energy side. The error bars in the figure reflect only statistical uncertainty.

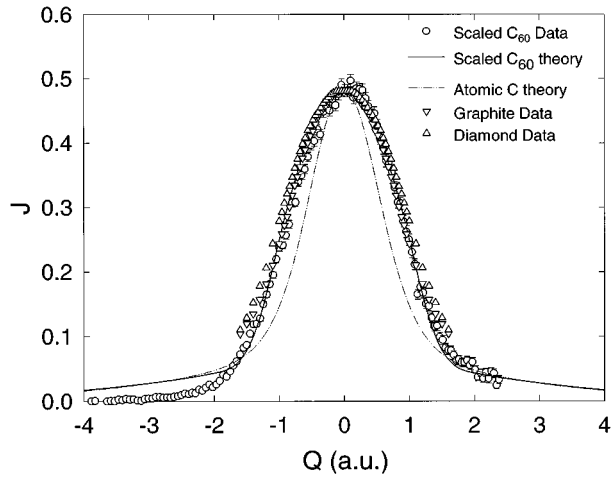

FIG. 3. Experimental and theoretical Compton profiles. The solid circles are from this work and represent the experimental Compton profile for C_{60}; the solid curve is the corresponding theoretical Compton profile. The remaining curves are from the literature. In order to facilitate comparisons, all Compton profiles were scaled to the atomic carbon profile at the peaks.

In order to facilitate comparison between theory and experiment, the experimental C_{60} Compton profile in Fig. 3 has been shifted by 0.21 a.u. of momentum. This shift is probably necessary due to the use of too small a value for E_I in eq. (2). As expected, using different values of E_I had the effect of shifting the Compton profile, without noticeably changing its shape.

For theoretical calculations of the C_{60} electron wave functions we have used a spherical model in which the determination of the Compton profile is straightforward. The model has been described[4] previously in detail. Briefly, the C_{60} molecules form a solid phase in which the molecules are bound together by a van der Waals-type interaction. Therefore the natural starting point for the calculation of the wave functions is the electronic structure of a single C_{60} molecule. In this model the valence electrons (four per carbon atom) move in a spherically symmetric field due to a spherical shell of uniform rigid positive charge and due to the spherical electron density. The radius of the shell is the radius of the C_{60} molecule and the thickness of the shell is determined by requiring charge neutrality. In order to reproduce the electronic properties, such as the widths of the occupied spectra of the σ and π states and the ionization potential, it has been found necessary to introduce an adjustable potential shift inside the shell of the positive charge.[4] The electron density is calculated self-consistently using the Kohn-Sham method of the density functional theory. Previously, this model has been shown to describe well the static polarizability and the photoabsorbtion of the C_{60} molecule.[4] We have made tests also by using a model (see e.g. Ref. 5) in which the shell of the positive charge is infinitely thin and there is no adjustable shift in the potential. The ensuing Compton profile is very similar to that obtained in the model described above reflecting its rather weak dependence on the parameters of the spherical model.

The occupied one-electron eigenstates in the spherical model consist of states with principal quantum numbers $n=1$ and $n=2$. The $n=1$ states have no nodes in the radial direction and correspond therefore to σ states. The $n=2$ states have one node in the radial direction and correspond to π states. The $n=1$ states are occupied from angular momentum $l=0$ to $l=9$, and $n=2$ from $l=0$ to $l=4$.

In order to test these wave functions we first convert them to momentum space and then generate a Compton profile using[26] the following relation:

$$J_{\mathrm{th}}(Q)=\sum_{nl}^{\mathrm{occ}} 2(2l+1)\int_{Q}^{\infty} p|\chi_{nl}(p)|^2 dp, \tag{3}$$

where the sum is over the occupied states and the momentum density $|\chi_{nl}(p)|^2$ of an electron in the nl shell is given by

$$\chi_{nl}(p)=\left(\frac{2}{\pi}\right)^{1/2}\int_0^{\infty} r^2 R_{nl}(r) j_l(pr)\,dr. \tag{4}$$

Here $j_l(pr)$ is a spherical Bessel function, and $R_{nl}(r)$ is the radial spatial wave function.

In order to have the momentum content of exactly 240 valence electrons of the C_{60} molecule we use in the sum of eq. (3), for the highest occupied level $n = 1$, $l = 9$ the degeneracy factor of 28 instead of $2(2l+1) = 38$. The momentum content of the carbon $1s$ core electrons was calculated[26] using the free atom core wave function and the contribution of these 120 core electrons to the Compton profile was added to the valence electron contribution.

The resulting theoretical Compton profile is shown in Fig. 3, along with the Compton profile[26] for atomic carbon, and the experimental[27] Compton profiles for C_{60}, diamond and graphite. For purposes of comparison, all Compton profiles were scaled to the atomic Compton profile at the maxima. The area under the atomic Compton profile is normalized to 1. The Compton profiles for diamond and graphite, which were measured using conventional Compton scattering,[27] are averaged over all orientations. Also, all of the profiles shown in Fig. 3 with the exception of the experimental C_{60} profile of this work, were reflected about $Q=0$. That is, tabulated values were only given[26,27] for $Q \geqslant 0$. Thus perfect symmetry is seen for those results, a symmetry not seen in the experimental part of this work.

It can be seen that the experimental and theoretical Compton profiles for C_{60} are virtually identical. The slight discrepancy on the $Q<0$ wing is probably due to the uncertainty incurred in the background subtraction. The theoretical Compton profile for atomic carbon is noticeably narrower than those for C_{60}. This may be interpreted to mean that the valence electrons are more localized in C_{60} than are the L-shell electrons in atomic carbon. From a comparison of the curves in Fig. 3 one sees that C_{60} more closely resembles graphite than either atomic carbon or diamond, insofar as the valence electrons are concerned. This is reasonable since the bonds in graphite are co-planar and, to the extent that a fullerene may be viewed as a curved 2-dimensional surface, so are the bonds in C_{60}.

To summarize, a novel technique was used to measure the C_{60} Compton profile. This Compton profile was compared with that for atomic carbon, graphite, and diamond, as well as a theoretical one for C_{60}. It is found that the experimental Compton profile for C_{60} is almost identical to the theoretical one, and is wider than that for atomic carbon, indicating that the electrons are even more spatially localized than in the atomic case. The near perfect agreement between theory and experiment is a necessary (but not sufficient) condition that has been met by the C_{60} wave functions.

The authors acknowledge Professor Uwe Thumm for several very helpful discussions, and Mr. James Norris for doing some of the preliminary computations of the theoretical C_{60} Compton profile. The experimental parts of this work were supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Basic Energy Research, U.S. Department of Energy.

[1] J. H. Weaver, J. L. Martins, T. Komeda, Y. Chen, T. R. Ohno, and G. H. Kroll, Phys. Rev. Lett. **66**, 1741 (1991).
[2] P. J. Benning, D. M. Poirier, N. Troullier, J. L. Martins, J. H. Weaver, R. E. Haufler, L. P. F. Chibante, and R. E. Smalley, Phys. Rev. B **44**, 1962 (1991).
[3] M. B. Jost, N. Troullier, D. M. Poirier, J. L. Martins, J. H. Weaver, L. P. F. Chibante, and R. E. Smalley, Phys. Rev. B **44**, 1966 (1991).
[4] M. J. Puska and R. M. Nieminen, Phys. Rev. A **47**, 1181 (1993).
[5] G. Wendin and B. Wästberg, Phys. Rev. B **48**, 14 764 (1993).
[6] A. Rubio, J. A. Alonso, J. M. López, and M. J. Stott, Physica **183**, 247 (1993).
[7] M. Cooper, Adv. Phys. **20**, 453 (1971).
[8] *Compton Scattering*, edited by B. G. Williams (McGraw-Hill, New York, 1977).
[9] M. J. Cooper, Rep. Prog. Phys. **48**, 415 (1985)
[10] N. Stolterfoht, in *Structure of Collisions of Ions and Atoms*, edited by I. A. Sellin (Springer, Berlin, 1978), p. 155.
[11] D. Burch, H. Wieman, and W. B. Ingalls, Phys. Rev. Lett. **30**, 823 (1973).
[12] H. Böckland and F. Bell, Phys. Rev. A **28**, 3207 (1983).
[13] D. Brandt, Phys. Rev. A **27**, 1314 (1983).
[14] H. I. Hidmi, C. P. Bhalla, S. R. Grabbe, J. M. Sanders, P. Richard, and R. Shingal, Phys. Rev. A **47**, 2398 (1993).
[15] D. H. Lee, T. J. M. Zouros, J. M. Sanders, H. Hidmi, and P. Richard, Nucl. Instrum. Methods Phys. Res. B **79**, 11 (1993).
[16] T. J. M. Zouros, P. Richard, and D. H. Lee, *High-Energy Ion-Atom Collisions, Lecture Notes in Physics 376*, Debrecen, Hungary, 1990, edited by D. Berényi and G. Hoch (Springer, Berlin, 1991).
[17] H. I. Hidmi, P. Richard, J. M. Sanders, H. Schöne, J. P. Giese, D. H. Lee, T. J. M. Zouros, and S. L. Varghese, Phys. Rev. A **48**, 4421 (1993).
[18] T. J. M. Zouros, D. H. Lee, J. M. Sanders, and P. Richard, Nucl. Instrum. Methods Phys. Res. B **79**, 166 (1993).
[19] B. D. DePaola, P. Richard, Y. Kanai, T. Kambara, and Y. Awaya, J. Phys. B **28**, 4283 (1995).
[20] R. Parameswaran, B. P. Walch, S. Maleki, C. P. Bhalla, and B. D. DePaola, Phys. Rev. A **47**, 3801 (1993).
[21] R. Parameswaran, Ph.D. thesis, Kansas State University, 1992 (unpublished).
[22] R. Parameswaran, W. J. Axmann, T. J. M. Zouros, and B. D. DePaola, Nucl. Instrum. Methods B **40**, 158 (1989).
[23] J. Abrefah, D. R. Olander, M. Balooch, and W. J. Siekhaus, Appl. Phys. Lett. **60**, 1313 (1992).
[24] Ulvick Industries, Inc., Houston, TX, USA.
[25] In general, the cusp is made up of electrons which have been captured from the target to the continuum of the projectile and electrons which have been lost by the projectile to its continuum. Here, since the projectile is bare, the cusp is made up solely of electron capture to the continuum.
[26] F. Biggs, L. B. Mendelsohn, and J. B. Mauer, At. Data Nucl. Data Tables **16**, 201 (1975).
[27] R. J. Weiss and W. C. Phillips, Phys. Rev. **176**, 900 (1968).