Improvements in the contemporary photoemission spectroscopy implementation: A message to the ARPES community

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Abstract

In this short communication, we highlight the deficiencies within the contemporary angle resolved photoemission spectroscopy (ARPES) implementations and point out few remedies towards their resolution. These deficiencies prohibit the current version of the ARPES technique from revealing the many-body physics of solids in its entirety. It is believed that the origin of these deficiencies lie within the prevailing data acquisition methods used for registering information from the photoelectron. It is argued that a slight change in the data acquisition methods would remedy the situation.

Discussion

Recently we have published a paper [1] highlighting the inadequacies of PES as a technique for studying the many-body physics of correlated electron systems. The paper mainly discusses these inadequacies by invoking fundamental quantum mechanical concepts like the wave function collapse phenomenon in PES as their origin. It is argued that the measurement of a single particle property (e.g. kinetic energy) in PES from the photoelectron causes its many-body wave function to collapse leading to a loss of the quantum information carried by the photoelectron. As a result the PES spectrum is unable to display the many-body physics in e.g. Kondo systems etc. This phenomenon of wave function collapse is similar to the one argued for the case of an Einstein-Podolsky-Rosen (EPR) measurement on an entangled electron pair [2]. After putting a deeper thought/scrutiny we realize that indeed the EPR phenomenon is at work in the contemporary version of PES implementation. Having had some experience working with the PES technique (primarily using VG Scienta electron analyzers [3]), we would like to offer some suggestions concerning the remedial measures for addressing these inadequacies. We sincerely hope that the ARPES community would take our suggestion seriously and do the necessary changes to the instrumentation and/or data acquisition procedures in order to restore the power of photoemission spectroscopy as a tool for studying the many-body physics in condensed matter.

The data acquisition mode normally used in the electron energy analyzers (e.g. VG Scienta electron analyzer) for recording the ARPES spectra is the ADC mode. In this mode the photoelectron hitting the microchannel plate (MCP) detector generates a
current pulse. This current pulse is further analyzed in order to determine the count rate. In all cases the output is ultimately determined in terms of count rate (counts per second - cps or total counts). It is this determination in terms of 'discrete' counts which causes the EPR [2] phenomenon since the detected electron has been 'identified' separately from others which causes its wave function to collapse. As a result we get information about the collapsed wave function for the electron when we measure the discrete counts (or count rate) - which is being traditionally done in PES worldwide using Scienta as well as other analyzers. At this juncture we suggest that the photoemission intensity measurement need to be performed directly in terms of the channel electron multiplier (CEM) analog output current and plotted vs. the photoelectron kinetic energy/binding energy. Such a spectral profile will manifest complete many-body physics of the electrons since the photoemission current measurement would not cause the wave function collapse (because the current measurement does not study any particular electron). In a sense a more appropriate word for this variant of ARPES would be angle resolved photoemission current spectroscopy (ARPCS) since we are registering the photoemission current directly instead of discrete photoelectron counts. At this point one might be tempted to think that the photoemission current is simply obtained from the recorded discrete count rate after multiplying it by the elementary electronic charge. However a crucial catch in this line of thinking is that the discrete counts are the electrons in the collapsed state each having its mass as the free electron mass, its charge as an elementary electronic charge etc.; on the contrary the photoelectrons during the photoemission current measurement would remain in an entangled/superposed state as such the effective masses for those electrons could be different from the free electron mass, their effective charges could be different from the elementary electronic charge and hence the current contributed by such charge carriers would yield a different value from that calculated from the discrete count rate. Thus the resulting ARPCS spectrum will be different from that obtained by recording the count rate (ARPES spectrum).

There could be different ways to determine the photoemission current from the CEM. We would prefer to make the photoemission current measurement (using a current meter) just analogous to the tunneling current measurement as done in tunneling spectroscopy. This involves a 'continuous' measurement which is made possible when using a continuous photon source. Normally the photon sources used in PES are indeed continuous and hence the above measurement scheme should be feasible in most of the cases. Another idea would be to record a digital equivalent of the amplitude of the current pulse at the output of the CEM by using analog-to-digital-converter and using this value to represent the photoemission intensity (please note that in the traditional versions of the data acquisition modes, every current pulse registers only one count irrespective of its amplitude which is already a substantial loss of quantum information). Thus in this new scheme we record a quantity directly proportional to the photoemission current amplitude so in some sense we are indeed measuring the photoemission current itself within the proportionality factor.

We hope that we have elaborately clarified our viewpoint on the deficiencies within the contemporary PES implementations and pointed out few directions towards its remedy. These remedial measures might lead to a development of a new variant of ARPES known as ARPCS which would be a revolutionary experimental technique for studying the many-body physics of the systems. Combined with high resolution capabilities of the current electron analyzers (e.g. VG Scienta analyzers etc.) this
mode of experimental measurement would offer great scope for progress in contemporary condensed matter research. We sincerely hope that our suggestions would make an impact over the future designs/upgrades of the electron analyzers.

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Appendix
Improvements in the contemporary photoemission spectroscopy implementation: A message to the ARPES community

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Abstract

In this appendix to the original article, we make an attempt to pinpoint the actual changes necessary to be made in the electron detection system employed in the electron analyzers within the ARPES instrumentation. This will provide a guideline on how to effect our idea into practice within an example case of the electron detection system used in Scienta Hemispherical electron analyzer.

Discussion

The idea regarding the changes to be made in the electron detection system in ARPES technique, which is presented in the main article associated with this appendix, will be demonstrated in this appendix taking an example of the electron detection system employed in Scienta Hemispherical analyzers [A1]. In Scienta analyzers the electron detection system consists of a multichannel plate (MCP)-Phosphor screen (PS)-CCD camera (CCD) assembly in which we measure the intensity of the ARPES signal, in terms of discrete counts, as a function of the electron’s kinetic energy and its angle of emission. MCP acts like an electron amplifier acting as an input to the PS for causing scintillations on the PS the intensity of which are then assigned digital values by the CCD camera and the associated electronics.

In the case of aforementioned detection system if you take any two photoelectrons from the sample (or for that matter any two electrons in the universe which are incident on the MCP!) and record the final digital values assigned by the CCD camera+electronics to them (after they create charge pulses upon impacting the MCP and subsequent scintillations on PS), they will always be identical and will be independent of the details of the many-body states from which the photoelectrons are emitted. This is a gross error/shortcoming of the electron detection system since the photoelectron under study could have highly renormalized properties inside the solid due to the many-body effect however the final digital value recorded for it is the same as that for another photoelectron which could be barely affected by that many-body effect. Ironically the study of such many-body effects is exactly our goal while using this technique. Thus we see that the electron detection system as mentioned above is incapable of studying the many-body effects in its entirety. For example, in case of ‘heavy’ fermion systems, the electron which is ‘heavy’ should contribute a
measurably distinct final digital value to the photoelectron intensity as compared to the electron which is not ‘heavy’. Only then we can say that this technique is really capable of studying the many-body phenomena of solids. Of course there are examples of some other many-body phenomena, e.g. BCS spectral function \([A2, A3]\), which can be studied reliably using this technique despite the deficiencies pointed out as above. But this is so because of a different reason that the exact nature of the many-body effect varies from one phenomenon to other and different many-body effects can have different impacts on the aforementioned electron detection system. In the above case, any many-body state of the sample corresponding to spin fluctuations of the localized spin inside the sample (the Kondo state being one of the examples of such a spin fluctuating state) cannot be studied using such an electron detection system.

{A ‘figurative’ description which will help you in understanding this concept is: The photoelectron when ‘measured’ in the detector has a well defined spin state (i.e. either up or down spin but not a superposition of both). Hence any many-body state of the solid which involves spin fluctuations of a localized spin inside the solid cannot be studied by this technique since the carrier of the quantum information of that spin fluctuating many-body state of the solid, i.e. the photoelectron, does not ‘preserve’ the spin fluctuations of that many-body state at the moment it gets ‘measured’ in the electron detector. As such this photoelectron is unable to ‘transfer’ the information of the many-body state from which it originated to the electron detector. Hence in the current version of ARPES implementation, due to the deficiencies in the electron detection mechanism, the many-body physics of the solids cannot be studied in its entirety. However rectifying the electron detection mechanism by measuring the electric current from the photoelectron would allow even a superposed spin state of the photoelectron during the measurement (which allows for a spin fluctuating many-body of the solid to be studied) hence the photoelectron is allowed to carry and transfer all of the information it collected from its original many-body state to the electron detector. This is how ARPES can be modified to tap its full potential.} 

Thus the problem can be remedied by modifying the electron detection system in such a way that the final digital value recorded by the detector for every photoelectron carries the ‘imprint’ of the nature of the many-body state from which it is emitted. By this we mean that the final digital value for every photoelectron, in general, should be distinct. How to ensure this in the aforementioned electron detection system? Well, the problem with the existing electron detection system can be identified as arising due to the process of converting the charge pulse from the output of MCP to light pulse by the PS. This process destroys the superposition/entanglement of the photoelectron arising from its many-body state as a result we have an identical output signal from the PS for every photoelectron incident on the MCP. This phenomenon results from the nature of the excitation-deexcitation mechanism (giving rise to the emission of photons causing scintillations) of the atoms in the PS (or in the solids in general) leading to the destruction of the superposition/entanglement (arising from the many-body state of the solid which has to be studied) carried by the incident photoelectron as a result of the wave function collapse. This is because the ‘optical gain’ (conversion factor of the PS i.e. the no. of photons produced per electron striking the PS) produced by every photoelectron (irrespective of whether it is a ‘heavy’ or ‘light’ electron) striking the PS is identical to each other since all the electrons have identical kinetic energies when they strike the PS as they are detected in constant pass energy mode (+ the PS screen voltage is kept identical for every electron) within the Scienta analyzer {In reality every photoelectron’s kinetic energy
is not strictly identical to each other but lies within the energy bandwidth allowed to pass through the analyzer at a time which is roughly 10% of the pass energy of the analyzer. The energies within this band are distributed across different microchannels and thereby along the energy axis of the detector. Thus every microchannel roughly corresponds to a unique kinetic energy for the photoelectron. When a ‘heavy’ electron and a ‘light’ electron passes through a particular microchannel then the detector pixel at the output of the microchannel is not able to distinguish between the two as they both strike the PS with same kinetic energy. Therefore it seems that the detector is insensitive to the details of the many-body information carried by the photoelectron. Thus these new ideas presented by us should be applied separately to each microchannel and to the detector pixels appearing immediately after its output in the case of simultaneous detection of photoelectrons within an allowed energy bandwidth. The identical final digital value for every photoelectron amounts to having a collapsed state for the photoelectron where every photoelectron seems to be equal to each other in its quantum information content! Otherwise, ideally, one should expect the intensity of the PS scintillation (arising from an incident photoelectron) and thereby the final digital value assigned to that scintillation, to vary across different photoelectrons due to the differences in the quantum information carried by different photoelectrons (concerning a variety of the many-body states of the solids.). Thus the remedy would be to remove the PS-CCD camera assembly completely and measure the electric current at the output of the MCP for each microchannel separately. The magnitude of the electric current for each microchannel reflects the photoelectron intensity for that channel. The measurement of the electric current would preserve the superposition/entanglement of the incident photoelectron as such its many-body information could be studied entirely through this measurement scheme. For example, the value of the measured electric current in case of the ‘heavy’ electron coming out of a heavy fermion/Kondo compound would be different than that measured for a ‘light’ electron. Other parts of the analyzer would remain the same. The instrumentation/electronics controlling the energy axis and angle axis dispersions of the analyzer should be kept the same as before.

{An issue which may arise in the reader’s mind here is that in our proposed modification of the detector we are allowing the kinetic energy selection to be done for the photoelectron whereas in ref. [A4], we had contested the measurement of the kinetic energy of the photoelectron. Actually there is no contradiction between both the pictures as in our modification of the detector we are not measuring the kinetic energy of a single electron at a time. The process of measurement of the electric current never allows an exclusive contribution to it by a single/specific electron. So when we are doing the kinetic energy selection and subsequent measurement of the electric current, all the electrons of the sample are partially contributing to the measured current at a time due to the superposition/entanglement resulting as a consequence of exchange symmetry between the photoelectron and the remaining electrons inside the sample. So it means that in this case we are not determining the kinetic energy of a ‘single’/specific electron at a time while in the aforementioned reference we contested the extraction of kinetic energy information from a ‘single’/specific electron. So our new picture as proposed in this appendix does not conflict with ref. [A4].}

Currently it is believed that the relative variation of intensities in the ARPES spectra detailing the many-body renormalization of the electronic states results from the statistics of the photoemission process in that, for example, more no. of ‘heavy’ electrons are emitted (as compared to the no. of ‘light’ electrons emitted) within a
definite time interval of the data acquisition process in order to generate the predicted narrow Kondo resonance peak in the experimental spectrum. However we beg to differ that there is an additional factor affecting the ARPES intensities coming from the many-body contribution embodied within the single particle spectral function \((\text{calculated using Green's functions, see ref. [A5]) denoted by } A^+(\mathbf{k}, \omega) \text{ and } A^-(\mathbf{k}, \omega)\) (one particle addition and one particle removal spectral function respectively) which is highly influenced by the quantum phenomena like e.g. Einstein, Podolsky, Rosen (EPR) \([A6]\) correlations between two electrons etc. Therefore the one particle spectral function as obtained in the collapsed state of the photoelectron wave function would be notably different than that in its uncollapsed state. Therefore this fact should be taken into consideration in the details of any experiment, e.g. electron detection mechanism in case of ARPES etc., in order to apply such a many-body theory to that experiment. In the theoretical derivation of \(A^+(\mathbf{k}, \omega)\) and \(A^-(\mathbf{k}, \omega)\) we 'implicitly' assume that the incoming electron towards the solid or outgoing photoelectron from the solid remains in its superposition/entanglement with the remaining electrons of the solid. Such a superposition/entanglement is a result of the antisymmetrization of its wavefunction with the wavefunction for the remaining electrons in the solid. As a result the comparison between the results of an experiment where such an antisymmetrization condition is violated during the experiment, e.g. the photoelectron detection process in case of the present-day version of ARPES, with a theory which implicitly assumes it, is inappropriate according to our view.

The photoemission spectrum is usually simulated within a sudden approximation to the perturbation imposed by the incoming photon in which the outgoing photoelectron is disconnected from the remaining solid entirely. Therefore it is assumed that there will be no energy exchanges between the photoelectron and the remaining solid post photoemission process. Therefore our claim of the loss of the antisymmetrization in ARPES electron detectors creating remarkable changes in the ARPES experimental spectra may sound scientifically illogical. However it is to be noted that the loss of superposition/entanglements corresponding to a particle is well known to have notable effects on the particle spectra/behavior in quantum mechanics (and these effects are in addition to the effects arising due to various approximations used in quantum theory like sudden/adiabatic approximation to perturbation theory etc. In fact they originate from the fundamental principles of quantum theory as such these effects play a dominant role in determining the single particle/many particle spectra/behavior. The validity of sudden approximation does not excuse the photoelectron’s wavefunction of its antisymmetry with respect to remaining electrons of the solid. Hence loss of the antisymmetry would certainly have visible effects in the photoelectron’s experimental spectrum. A classic example in this regard is the Young’s double slit experiment with electrons. It is well known how the electron pattern on the screen kept beyond the double slit gives remarkable redistribution of electron intensity when the superposition of the incident electron is lost by forcing the electron to pass through only one slit out of the double slit at a time. All these changes are purely due to the quantum mechanical effects arising out of the destruction of the superposition but not due to strong Coulombic interactions etc. between electrons. In analogy, a striking change seen in the photoelectron’s experimental spectrum upon the loss of its superposition/entanglement with the remaining electrons of the solid should not come as a surprise given our knowledge of the peculiarity of the quantum phenomena around us. Incidentally, such a change has no bearing on the validity/invalidity of the sudden approximation applied to the photoemission process.
(or the validity/invalidity of the 3-step or 1-step model for the photoemission process). The antisymmetrization of the photoelectron's wavefunction with the wavefunction of the remaining electrons of the solid holds true in all the models or approximations used for describing the photoemission process i.e. 3-step or 1-step modeling of the photoemission process OR sudden or adiabatic approximation applied to photon induced perturbation etc. and as such the photoelectron’s experimental spectrum will be influenced by the quantum effects arising due to the loss of the antisymmetrization of its wavefunction irrespective of the models/approximations used for describing the photoemission process.

There are two possibilities for measuring the electric current from the photoelectrons. One can either measure it before the MCP or after the MCP. We think that these two measurements would give an identical ARPCS spectrum (let us call the experimental spectrum obtained by measuring the photoelectron current instead of discrete photoelectron counts as ARPCS spectrum in order to avoid confusion with usage of the word ARPES spectrum), except for the amplification achieved by the MCP due to its gain factor. But this fact can actually be verified in an experiment by comparing the ARPCS spectra obtained in both the cases. A word of caution concerning the electric current measurement before/after the MCP is about the technique/instrument used for measuring it. The necessity is that we need to preserve the superposition/entanglement of the photoelectron in this technique, right from the moment the photoelectron was created till the final moment when the quantum information is extracted from it, in order for it to reveal the information about the many-body physics. This is a very delicate issue in that a slight mistake/inaccuracy may result in the superposition/entanglement of the photoelectron getting destroyed. Hence utmost care should be taken in choosing those current measurement methods/technologies which will not destroy the superposition/entanglement. For instance, we already discussed that converting charge pulses to light pulses by the PS was an incorrect method of measuring the ARPES intensity. We prefer the ammeters (based upon Operational Amplifier (OpAmp) based current-to-voltage converter) used in the tunneling spectroscopy experiments \[A7\] to be adapted here for the measurement of the electric currents of each microchannel separately. These ammeters have been successfully performing within the tunneling experiment and have yielded the reputation to the tunneling spectroscopy as a successful technique for measuring the many-body physics.

While measuring the electric current before the MCP, one should be wary of the fact that the electric currents would be extremely feeble (as the no. of photoelectrons appearing at the detector before the MCP would be extremely small to give any significant amount of electric current) and would require sensitive instruments to measure such small currents. In order to measure this current we propose to create a rigid two dimensional array of micrometer size (diameter) metallic charge collectors (similar to the ones which are used post channeltron amplifiers for collecting the resultant electron charge cloud from the output of the channeltron, except for their diameters which should be scaled to match roughly the diameter of a microchannel) arranged side-by-side to each other (just like the different microchannels of the MCP) and have this array replace the MCP. Thus this array would also be positioned within the \(E-\theta\) plane \((E \rightarrow \) kinetic energy of the photoelectron and \(\theta \rightarrow \) angle of emission of the photoelectron\) just like the MCP was positioned earlier. Then the electric currents due to the photoelectrons collected by individual charge collectors should be
measured individually by the aforementioned OpAmp based current-to-voltage converter circuit. The value of this electric current will give the intensity of the ARPCS spectrum corresponding to that point in the $E$-$\theta$ plane occupied by the charge collector (note that due to the existence of $E$ and $\theta$ dispersions of the photoelectrons along the perpendicular and parallel directions of the analyzer slit respectively, different charge collectors will measure the ARPCS intensity at different points in the $E$-$\theta$ plane simultaneously. Thus we will be able to map the (many-body effect induced) dispersions in $E$ and $\theta$ for the photoelectrons through this measurement scheme.). However the current-to-voltage converter must be tuned to measure very small values of current (picoampere or below) since the photoelectron currents would be very feeble in the absence of MCP.

The case of the electric current measurement post MCP is relatively easier. One needs to place these metallic charge collectors at the end of each microchannel separately and then measure the currents flowing through these charge collectors individually using the aforementioned OpAmp based current-to-voltage converter. However the current-to-voltage converter, in this case, should be tuned to measure currents of the order of microampere which normally come out of the microchannels of the MCP post amplification.

{Note: Recently we learnt about a new technology of IonCCD based charged particle detectors [A8] which can be used as an electron detector post MCP without the need for converting the electron pulse into a light pulse. Thus, apparently, this technology also appears to be promising in correcting the deficiencies in the electron detection system of ARPES. This technology may come in handy in replacing the PS-CCD assembly (both IonCCD and the photosensitive CCD used in the PS-CCD assembly are examples of pixelated anodes and hence would form an easy substitution between them) in the existing ARPES setup thus proving to be light on the changes to be made to the hardware of the electron detection system of the present-day ARPES, during the correction. We are exploring the details of this technology further in order to study the adaptability of the IonCCD based electron detector with regards to its two-dimensional position sensitivity of the electron detection as required by the technique of ARPES/ARPCS, as well as the exact physical processes involved in the charged particle detection and the exact details of the signal processing needed post charged particle detection in order to address the deficiencies of the electron detection system of present-day ARPES.}

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