Practicing DSAM in aberrant domain: use of multi-disciplinary techniques

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Abstract.
Measurement of level lifetime of nuclear states is of relevance in nuclear structure research as it provides us with an unique probe into the underlying microscopic structure of these states. Of the several experimental techniques for lifetime measurements, the Doppler Shift Attenuation Method (DSAM) is the one adopted for measuring lifetimes typically in the range of few tens of fs to few ps. The technique is based on the analysis of the observed Doppler affected gamma rays emitted by the recoils in flight. The crucial component in the related analysis is the simulation of the stopping process, of the residues of interest, in the target and the backing media. This requires calculation of the corresponding stopping powers and the same has been identified as one of the principal uncertainties in the extracted lifetime in DSAM. Traditionally the method is pursued with a thin target, for production of nuclei of interest, on a thick elemental backing wherein stopping process is perceived to occur. The present work in light of it’s objectives uses a setup which is in sharp variance with the conventional scenario, such as the use of a thick molecular target, which contributes both to the production of the residues as well as their subsequent slowing down. This demanded extensive developments in the analysis procedure particularly in the domain of simulating the stopping process with due incorporation of the nuances of nuclear reaction kinematics besides subjecting the molecular medium to a detailed structural characterization, routinely carried out in the domain of material science. These developments have been used to extract the level lifetimes of nuclei at the interface of the sd & pf shells such as ²⁶Mg, ²⁹Si, and ³²P.

1. Introduction
The nucleus provides us with a unique laboratory to investigate the two extreme degrees of freedom viz. single particle and collective. One of the primary objectives of any contemporary experimental nuclear structure endeavour is to populate high angular momentum states in the nucleus of interest, using an appropriate nuclear reaction, and study its de-excitation through emission of γ-rays that are detected using large arrays of high resolution detectors supported by state-of-the-art pulse processing electronics and data acquisition systems. Such efforts lead to detailed information on the level structure (excitation pattern) of the nucleus of interest that
includes the energies, spin-parities, and lifetimes of the states along with the \( \gamma \)-ray transitions connecting them. These are eventually compared with the model calculations for interpretation of the underlying degrees of freedom based on the predicted microscopic configurations.

The lifetime of a level encodes information on its microscopic structure (wavefunction) and the overlap with other available states, thus providing a stringent constraint on the model calculations. Measurement of level lifetimes is, thus, of much significance in nuclear structure research and is zealously pursued in many of the related endeavours in the domain. The experimentally measured lifetimes and the transition probabilities extracted therefrom are often compared with the theoretical predictions to validate the model interpretation of the level structure. Several procedures \[1\] of lifetime measurements have been practiced for decades and the choice of the same depends on the range of lifetimes being addressed in a particular investigation.

The Doppler Shift Attenuation Method (DSAM) is adopted for measurement of level lifetimes in the range of \( \sim \) few tens of fs \((10^{-15} \text{ s})\) to \( \sim \) few ps \((10^{-12} \text{ s})\). Nuclei of interest produced in a reaction recoil through the target and the backing media, slowing down and eventually stopping in the process. In conjunction, these recoiling nuclei also de-excite from the state they are initially populated (entry state) through the characteristic level sequences, by \( \gamma \)-ray emissions, finally into the ground state. If the lifetime of a level is less than or of the order of the aforementioned stopping time, the de-exciting \( \gamma \)-ray from the level may be emitted from a recoiling nucleus in-flight and exhibit Doppler effect, observed in the resulting spectrum, as per the equation,

\[ E_{\gamma}(\theta) = E_{\gamma0}(1 + \beta \cos \theta) \]  

where \( E_{\gamma0} \) is the actual energy, \( E_{\gamma}(\theta) \) is the Doppler affected energy of the \( \gamma \)-ray, \( \beta \) is the velocity of the recoil during emission, and \( \theta \) is the angle of observation. In case of very short lived levels (lifetime \( \ll \) stopping time) and detector at angles \( \neq 90^\circ \), the \( \gamma \)-ray transition peak in the spectrum is shifted, compared to the actual energy, to higher (lower) energies when observed at forward (backward) angles with respect to the direction of the recoiling nuclei (close to the beam direction). For levels with lifetime \( \lesssim \) stopping time, the peak has a stopped component at the actual energy and an accompanying shape, extending to higher (lower) energies for detectors at forward (backward) angles. Typically observed Doppler shapes and shifts in the \( \gamma \)-ray spectrum, obtained in the present work, are illustrated in Fig. 1. The DSAM pertains to analyzing these shapes and shifts to extract lifetimes of the respective levels.

The cardinal components of analyzing data from a DSAM experiment are as follows.

(i) The slowing down and stopping of the recoils in the target and the backing media is simulated using the Monte Carlo techniques. This requires, apart from other inputs, stopping powers of the target and the backing for the recoils of interest. Results of the simulation are typically the velocity \((\beta)\) and the trajectory information \(( \text{direction cosines})\) of a chosen number of recoils, in uniform time steps, starting from the production of the recoils through their slowing down and eventual stopping in the medium.

(ii) The velocity profiles of the recoils, as viewed by the detectors at different angles, are calculated using the results of the above simulations and an input of the detector geometry, such as the detector radius and the target-to-detector distance.

(iii) The expected Doppler shape / shift of a given \( \gamma \)-ray transition at different angles is calculated using inputs on the level structure, such as the energy of the transition being analyzed, initial estimate of the level lifetime, and side feeding description (intensity, feeding time) of the level. This is combined with the velocity profiles as viewed by the detectors.
Figure 1. Typical Doppler shape and shift observed in $\gamma$-ray spectrum following a DSAM measurement reported in the present paper.

...at different angles, from the preceding step, along with the information on the intrinsic resolution of the detector, to generate the calculated Doppler affected shaped/shifted $\gamma$-ray transition peak.

(iv) The calculated shape is fitted to the experimental spectrum through least square minimization, in order to extract the level lifetime of interest. The crucial parameters of fitting are essentially the level lifetime and the side feeding time.

The principal uncertainties in the DSAM analysis have been identified to stem primarily from (i) the estimation of the stopping powers that govern the slowing down process of the recoils and (ii) the side feeding models adopted in the analysis. The uncertainties owing to the side feeding can, in principle, be eliminated through use of gates set on transitions above the level of interest [5]. Thus, the major source of uncertainty in lifetime determination using the DSAM may be attributed to the knowledge of the stopping powers. The problem is further compounded in case the measurement requires deploying molecular target and backing wherein the estimation of stopping powers, using theoretical models, is even more difficult.
Of the several codes used for analyzing data from DSAM measurements, the LINESHAPE package [2] is one that is extensively used in many of the contemporary endeavours. Stopping powers are calculated using one of the earlier models by Ziegler [3] or Northcliffe and Schilling [4] that are based on the proton and α stopping data and uses different scaling algorithm for the heavy-ions. The stopping simulation for the recoils in the target and the backing is carried out by the DECHIST program of the LINESHAPE package, using Monte Carlo techniques. The approach holds good for a conventional DSAM measurement that customarily uses a thin target on a thick elemental backing so that the energy loss of the recoils in the target can be neglected and the stopping process can be perceived to be occurring entirely in the backing medium wherein the stopping powers, calculated from the above models, are reasonably accurate. Certain lifetime measurements, however, require setups that are incongruent with the traditional scenario and uses target that are molecular in nature. Moreover, depending on the method of target fabrication, the molecular target may be substantially thick so as to operate both as target as well as backing which, owing to the molecular nature, is beyond the description allowed by the conventional LINESHAPE-based analysis.

The neutron rich $^{18}\text{O}$ target, extensively used in the spectroscopy of $sdgf$ nuclei [5, 6], come in the form of metallic oxides, such as Ta$_2$O$_5$. The present work reports developments and modifications pertaining to the analysis codes and methodology for application of DSAM in such unconventional setup. The nuclei of interest in this work were populated in a series of experiments using the $^{18}\text{O}(^{18}\text{O},x_{pynz}\alpha)$, $^{18}\text{O}(^{13}\text{C},x_{pynz}\alpha)$ and $^{18}\text{O}(^{16}\text{O},x_{pynz}\alpha)$ reactions at $E_{lab} = 30-34$ MeV. The beam was provided by the Pelletron-LINAC Facility at the Tata Institute of Fundamental Research (TIFR), Mumbai and the Pelletron Facility at the Inter University Accelerator Center (IUAC), New Delhi. The neutron-rich $^{18}\text{O}$ target was prepared by heating a Ta foil of thickness $\sim 50 \text{ mg/cm}^2$, in an enriched $^{18}\text{O}$ atmosphere, resulting in the formation of Ta$_2$O$_5$ layer on either side of the foil. The thickness of the Ta$_2$O$_5$ on either side of the foil was estimated to be 9.25 mg/cm$^2$ which was sufficient to stop the recoils. For the experiment with the $^{18}\text{O}$ beam, an array of seven Compton-suppressed Clover detectors was used as the detection system while that in the experiments with the $^{13}\text{C}$ and the $^{16}\text{O}$ beams, it was the Indian National Gamma Array (INGA), then consisting of 15-16 Compton-suppressed Clovers. Further details on the experimental setups can be found in Refs. [5, 6].

The acquired data were sorted into angle dependent $\gamma-\gamma$ matrices for lifetime analysis, using the LINESHAPE [2] package. However, as indicated earlier, the programs / sub-programs of the package employed for simulating the slowing down process of the recoils, were replaced as detailed in the next section.

2. Modifications of the LINESHAPE Code for Thick Molecular Targets
The conventional LINESHAPE program does not allow for molecular media in simulation of the stopping process of the recoils in the target and the backing. Moreover, as indicated above, it is suited to a thin-target-on-thick-backing setup wherein the energy of the beam can be assumed to remain largely constant throughout the target thickness and consequently, the yield of the residual (recoiling) nuclei is uniform therein. The DECHIST program within the LINESHAPE code is used for these stopping simulations and it was modified to incorporate the characteristics of the thick molecular target setup.

Updated stopping powers for heavy ions in elemental as well as molecular media are available in contemporary software like SRIM [7], which have been exhaustively benchmarked with
Figure 2. (Color online) Stopping powers of $^{32}\text{P}$ ions in Ta$_2$O$_5$ using SRIM (labelled as SRIM in the legend), DECHIST with SRIM input and interpolation (DECHIST_SIRM) and DECHIST with effective Z and effective A for the Ta$_2$O$_5$ target (DECHIST_EFFECTIVE-Z).

Figure 3. (Color online) Cross-section of the residue as a function of beam energy. The original DECHIST program assigns uniform cross section (red line) of the residue while the modified DECHIST program incorporates beam energy dependent cross section (blue line) along the target thickness.

respect to the experimental data and are considered to be more reliable. The stopping powers of $^{32}\text{P}$ ions, one of the nuclei studied in these investigations, in Ta$_2$O$_5$ from SRIM are included in Fig. 2. In the light of the reliability of the SRIM stopping powers, these have been incorporated in the modified DECHIST program. The DECHIST program calculates the stopping powers at 61 hard-coded energies, ranging from 1 keV to 1000 MeV, for the residue in the target and the backing, as well as for the beam in the target. The stopping power required for all the intermediate energies are calculated through an in-built interpolation subroutine. In the modified version of the program the user is required to provide these stopping powers, calculated from the SRIM code, at these 61 energies through an input file. The interpolated stopping powers for $^{32}\text{P} + \text{Ta}_2\text{O}_5$ case, as calculated by the modified DECHIST program, are also presented in the Fig. 2 (DECHIST_SIRM). Nearly complete overlap between the stopping powers calculated by SRIM and DECHIST, with SRIM inputs and appropriate interpolation, validates the modification. Fig. 2 also illustrates the stopping powers calculated using an effective Z and an effective A to represent the molecular stopping medium in the conventional DECHIST program. The deviations in the stopping powers, calculated using the latter, at intermediate and higher energies indicate unacceptability of the approach.

Use of a thick target results in a substantial change (decrease) in the energy of the incident projectile as it traverses the target thickness and thus affect the production cross section of the residues along the depth of the target. Further, in a thick target, one needs to determine the thickness of the target that actually contributes to the residue production while the rest of the target thickness acts only as the stopping medium. The knowledge of target thickness contributing to the residue production follows from the information on the cross section depen-
dence on the beam energy and the evolution of the latter along the target thickness. The original version of the DECHIST program, which simulates the stopping of a given number of recoiling residues, divides the target thickness into bins equal to the (user) given number of recoil nuclei and treats each bin as the origin of one recoil nucleus. This is a valid approximation for a thin target through which the beam energy can be assumed to remain unaltered and thus produce the residues with uniform probability (cross-section). In the modified version of the DECHIST program used in the present work, changes were implemented to incorporate the cross-section dependence of the residues on the changing beam energy along the thickness of the target. In this version, up to 10 bins can be defined along the target thickness and relative cross-sections for the residue, in arbitrary units, can be specified therein, as illustrated in Fig. 3. It is preferable to specify the cross sections with reference to available experimental data or, in its absence, from statistical model calculations. It follows that the total thickness of the aforesaid bins is the thickness of the target that contributes in the production of the residues while the remaining thickness contributes only to the stopping process.

The use of molecular target introduces one more degree of uncertainty in the analysis. Most of the molecular targets are prepared as oxides and it is possible that these oxides appear in several structural phases. Some of these phases may also occur as a consequence of the temperature dependent preparation technique. Therefore, it is imperative to carry out a detailed structural evaluation of the target using well-established structural evaluation techniques such as X-Ray Diffraction (XRD) and Field Emission Scanning Electron Microscopy (FESEM) as detailed in Ref. [5]. These techniques were applied to the Ta$_2$O$_5$ target used in the present work and they revealed the presence of multiple crystalline phases, coupled to an amorphous phase in the sample. The SEM images of the Ta$_2$O$_5$ target are presented in Fig. 4. The presence of multiple phases would manifest as a modification in the density of the target medium, that will have a significant impact on its stopping characteristics. The consistent analysis of several known level lifetimes in a host of nuclei populated in the experiments indicated that a density of 5.76 g/cm$^3$ produces the best agreement with the reported lifetimes and hence this value has been used in the subsequent analysis.
Representative fits to the observed Doppler shapes of the 1677 and 2418 keV transitions in the $^{32}\text{P}$ nucleus, studied in the present work, using the modified LINESHAPE code, are illustrated in Fig. 5. The lifetime obtained for the 1755 keV level, de-exciting by the 1677 keV transition, is $616^{+65}_{-46}$ fs that is in excellent overlap with the literature value of $620^{+40}_{-40}$ [9]. An upper limit of $629^{+82}_{-61}$ fs was obtained on the lifetime of the 5862-keV level, de-populated by the 2418-keV transition and was reported for the first time [5]. The compliance [5, 6] of the lifetimes following the present analysis with those reported previously essentially validates the present procedure.

3. Methodology for inclusion of TRIM simulations in lifetime analysis

It may be noted that even though the stopping powers in the preceding analysis were being calculated through the SRIM code, the Monte Carlo simulations of the stopping process was still dependent on the DECHIST program of the LINESHAPE package. This might be replaced with contemporary stopping simulation codes such as TRIM that uses SRIM calculated stopping powers and updated statistical algorithms to simulate the transport of ions through material media. Use of such state-of-the-art simulation tools is also expected to render immense flexibility in the definition of the target and / or backing media that may be elemental or molecular, thin or thick, single or multi-layered, as allowed by the modern simulation packages such as TRIM.

The TRIM program, acronymed for TRansport of Ion in Matter, finds extensive use in the domain of material science research wherein it is applied to investigate the effects of energetic ions passing through a medium that might be elemental or molecular and might consist of mul-
tiple layers of varying depths. It is a common understanding that the code was never intended for use in lifetime analysis. It is an efficient simulation tool through use of statistical algorithms and thus is an alluring option for simulating the slowing down process of the recoiling nuclei in the target and the backing media as is required in analysis for lifetime measurements using DSAM.

To begin with, we consider a thick elemental or molecular target operating as the target, for production of the nuclei of interest, as well as the stopping medium. Our objective is to simulate the stopping process of the recoiling nuclei using the TRIM code and use the results of the simulation for subsequent analysis. The output of the TRIM simulations, however, cannot be naively used for DSAM studies. A set of programs have been developed for elaborate processing of the TRIM generated output to conform to the requirements of lifetime analysis. These pertain to correcting the trajectories with the reaction kinematics associated with the production of the residue, extracting the velocity profile (variation of $\beta$ with time) of the residues moving in the target-backing media, and incorporating the possible changes in the cross section for residue formation along the target thickness. Various aspects of the exercise are detailed in this section.

The TRIM code characteristically assigns a common starting point for all the simulated trajectories. The trajectories are all assumed to start from the origin (0,0,0) on the first surface of the stopping medium. The actual experimental scenario of lifetime measurements with thick target is, however, different. In case of a thick target, the incident beam traverses the thickness of the target and looses energy in the process. Consequently, the cross section for production of the residue of interest evolves along the thickness of the target and a varying number of residues are produced at different target depths, each with different starting points for their respective trajectories. In the current analysis, the cross section dependence of the residue of interest on the beam energy was determined either based on available experimental data or from statistical model calculations. This was combined with the calculations for change in the beam energy, along the target depth, using the SRIM code. It is understood that for a thick target, only a part of the same would contribute to the production of the residue of interest while the remaining thickness would operate only as the stopping medium. The effective thickness of the target, contributing to population of the residue, can be divided into bins representing the changing beam energy and the evolving cross section therein. Upto ten such bins can be considered in the present scheme. Each bin is characterized by a thickness (width), a beam energy and a yield (arbitrary units) of the nucleus being investigated.

The residues produced in each bin would have different initial energy-angle distribution and different starting points for their trajectories. The distribution in the initial energies and angle of the residues follows from the evaporation of the particles ($p, n, \alpha$), de-exciting the compound nucleus (Fig. 6) and has to be incorporated at the starting point of the trajectories in the stopping medium. The trajectories obtained from TRIM simulations, however, initiate aligned to the X-direction, with direction cosines (1,0,0), and does not include this distribution. The two-dimensional energy-angular distribution of the residues was calculated using the statistical model code PACE4 [8] for each beam energy corresponding to the individual bins, along the depth of the target. The number distribution of the residue in the energy-angle system, obtained from the statistical model code, is converted into a probability distribution for subsequent use.

The (stopping) simulation using the TRIM code is carried out for a residue energy greater than the highest possible energy, indicated by the statistical model calculations, typically for 10000 ions. The results of the TRIM simulations appear as trajectory information, in the form of the energy of the recoiling nuclei and the corresponding co-ordinates, recorded at certain
Figure 6. (Color online) Typical energy-angle distribution of residues generated in the statistical model calculation [8]. Schematic representation of a nuclear fusion-evaporation reaction producing residue (of interest) following particle evaporation from the compound nucleus. The TRIM trajectories, initiating aligned to the X-direction are rotated to match the angle of the residue chosen under the constraint of the energy-angle distribution.

energy interval. This output file containing the simulated trajectories serves as one of the input to our program. The program also reads-in, for each of the aforementioned bins, the respective energy-angle probability distribution file, relative yield of the residue (normalized to 10000) and the distance (in Angstrom) of the geometrical center of the bin from the surface of the target.

During execution of the program, an energy and an angle is chosen for each residue under the constraint of the probability distribution of the respective bin. The corresponding TRIM trajectory, chosen sequentially from the set of trajectories generated with identical incident energy of all the ions, is inspected for the point where the energy of the residue is nearly equal (or just lower than) to the chosen energy from the distribution. This point in the trajectory is then treated as the origin of the trajectory of this particular residue in the stopping medium. The origin is appropriately offset to initiate at the center of the respective cross section bin. All the subsequent points in the trajectory are subjected to the same offset. In conjunction, the angle chosen from the distribution is used to modify the trajectory through application of a rotation matrix, so as to incorporate the angular entry of the residue, deviated from the beam direction (following particle evaporation), into the stopping medium. The rotation aligns the initial direction of the simulated trajectory to the direction of the residue. The same rotation is subsequently applied to all the direction vectors in the simulated trajectory.

The stopping simulations for lifetime analysis are typically in the form of trajectory information recorded in uniform time steps. A given trajectory (history) is logged in terms of the $\beta$ and the direction cosines at each time step, the size of which is specified by the user and is chosen to be much less than the minimum lifetime being expected. The history is tracked from the initial $\beta$ to the point where the $\beta$ drops by 1/1000 of the initial value. The trajectory information obtained from TRIM and modified with inclusion of reaction aspects thus needs to be converted into a description wherein a trajectory is represented by the time profile of $\beta$ and the direction cosines, logged in uniform time steps, in compliance with the requirements of lifetime analysis. This conversion is implemented by considering the consecutive points in a trajectory, to calculate the change in the energy and the accompanying intermediate distance, from which the associated time is extracted. This is followed by re-binning the change of the $\beta$ and the direction cosines, between the consecutive points, in uniform time steps, through linear interpolation. The latter is a reasonable approximation owing to the small change in energy
(typically ∼ 1%) between the consecutive points in the record of the simulated trajectory. The size of the time step is user specified and is usually ∼ 0.001-0.002 ps.

In case of the more conventional scenario of a thin target on a thick backing, the working with the TRIM simulations is slightly different from that described above. This is particularly relevant if the residues loose an amount of energy in the target medium, before reaching the backing, that is non-negligible. In this case, the TRIM simulation is separately executed for the target and for the backing. Modified versions of the above program are used to individually process the simulation outputs for the target and the backing media. The program for the target uses the energy-angle probability distributions for the residues, generated using the PACE4 code, and produces, apart from trajectory info in terms of the β and direction cosines, a file containing energy and direction information for the trajectories at the target boundary. This file is read-in by the program to process the TRIM simulations for the backing medium. The energy and the direction of each residue at the target boundary is used by this program to modify the TRIM simulated trajectories through the backing medium and generate uniform time-binned β and direction cosines for these trajectories. The processed trajectories in the target medium and the backing medium are then combined in a common file. It may be noted that there may be trajectories in the target medium, particularly for not-very-thin target, that do not make it to the backing. Such trajectories are separately recorded and eventually combined with the common file with all the trajectories.

The output of this exercise are simulated trajectories, using experimentally benchmarked stopping powers from the SRIM database and state-of-the art Monte Carlo algorithms of the TRIM code with due incorporation of the details of the nuclear reaction and associated kinematics. This output is used for the subsequent analysis for extracting the level lifetimes. In the present work, the output of the stopping simulations is read-in by the HISTAVER program of the LINESHAPE package, to calculate the velocity profiles as viewed by the detectors at different angles. The subsequent steps for determining the level lifetimes are similar to the standard practice detailed in Ref. [5]. It should also be noted that the LINESHAPE package is being referred here in a very generic sense. The result of the stopping simulations using the TRIM code can well be made compatible with any code for DSAM analysis that may use these trajectory information for determination of level lifetimes.

Fig. 7 illustrates the representative fits to the Doppler shape / shift obtained through use of TRIM simulated stopping trajectories in the $^{34}$P and $^{26}$Mg nuclei. The corresponding lifetime results for the 3352-keV level, de-excited by the 1047-keV transition, in $^{34}$P is an upper limit of 475 fs that is consistent with the previously reported lifetime of $^{320}_{-115}^{+173}$ fs [9]. For the 1274-keV transition, de-populating the 3083-keV level, in the $^{26}$Mg nucleus, the lifetime results from the present analysis is an upper limit of 1339 fs, and determined for the first time in this investigation.

4. Conclusions

The present work attempts to address the uncertainties in the stopping powers used in the lifetime measurements with DSAM and extend its application beyond the conventional thin-target-on-thick-backing setups to the atypical thick molecular targets. The latter, as it is known, is indispensable in the spectroscopic pursuits of certain nuclei such as those in the $A$ ∼ 30 region, around $sd_{pf}$ shells, that require $^{18}$O target in the form of metallic Ta$_2$O$_5$ for investigations of their level structure. Stopping powers calculated from the updated and experimentally validated SRIM code has been used to analyze the Doppler shape / shifts observed in the spectra of these nuclei populated using heavy-ion induced fusion-evaporation reaction. While in the first step
Figure 7. (Color online) (Left Panel) Fitted Doppler shape of the 1047-keV transition, deexciting the 3352 keV level, in the $^{34}$P nucleus. (Right Panel) Fitted Doppler shape of the 1274-keV transition, de-exciting the 3083 keV level, in the $^{26}$Mg nucleus.

These stopping powers were applied within the existing code for DSAM analysis, LINESHAPE, in a further improvement the entire stopping simulation was carried out with the TRIM code using the stopping powers calculated by SRIM, and using the LINESHAPE code only for the subsequent analysis. The latter development is expected to extend the applicability of the DSAM technique to a wider range of target and/or backing media, as may be required for specific lifetime measurements.

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