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Abstract

A modified binary collision approximation (BCA) was developed to allow explicit evaluation of the times at which projectiles in a collision cascade reach significant points in their trajectories, without altering the "event-driven" character of the model. The modified BCA was used to study the temporal development of cascades in copper and gold, initiated by primary atoms of up to 10 keV initial kinetic energy. Cascades generated with time-ordered collisions show fewer "distant" Frenkel pairs than do cascades generated with velocity-ordered collisions. In the former, the slower projectiles tend to move in less-damaged crystal than they do in the latter. The effect is larger in Au than in Cu and increases with primary energy. As an approach to cascade nonlinearities, cascades were generated in which stopped cascade atoms were allowed to be redisplaced in later encounters. There were many more redisplacements in time-ordered cascades than in velocity-ordered ones. Because of the additional stopping introduced by the redisplacement events, the cascades in which they were allowed had fewer defects than occurred otherwise. This effect also was larger in Au than in Cu and larger at high energies although most of the redisplacement encounters involved only low-energy particles.

1. Introduction

The explicit temporal representation of the trajectories of atoms in collision cascades is widely ignored in simulations based on the binary collision approximation (BCA). Collisions in the trajectory of any one atom occur in the proper time order, but the cascade as a whole develops rather arbitrarily. MARLOWE [1–5] chooses the atom with the highest velocity as the projectile in each collision. Other programs use other rules: for example, at each collision TRIM.SP [6] follows the displaced target from the preceding collision if there was one or the projectile in that collision otherwise. Insofar as the model is within the scope of linear cascade theory [7], the order of the collisions is unimportant, but correlations involving the temporal aspects of cascade evolution cannot be evaluated if time is ignored nor can pictures of the development of cascades be produced. Finally, the possible importance of cascade nonlinearities cannot be assessed.

A method has been described for including time explicitly in BCA calculations without altering the "event-driven" character of the model [8]. The present paper shows how a BCA model with time-ordered collisions may be defined and how some aspects of collision cascades in static monocrystals of Cu and Au compare in time-ordered and in velocity-ordered calculations. In addition, the subject of cascade nonlinearities is approached by examining how often stopped cascade atoms become targets in later collisions.

2. The computational model

Time is included in MARLOWE by using the constant velocity of the barycenter in a collision as a clock. Besides adding a calculation of the time of each collision, to avoid temporal inconsistencies, extensive changes were made in the procedures for choosing appropriate target sites [8]. The aspects of MARLOWE that are important in the work reported here are described below. The references [1–5,8,9] should be consulted for further details.

2.1. Collision ordering

Each atom in a MARLOWE cascade is assigned a serial number. If the velocities are used to order the collisions, the serial numbers of atoms emerging from an encounter are put into a table at places based on the idea [10] that the collision density in a cascade varies roughly as E^{-2}, with E the kinetic energy of the atoms. The leading entry in this table is chosen for each collision. The algorithm accounts for differing atomic masses. Conflicts can occur between atoms for places in the table, so the algorithm is approximate.

If the collisions are to be ordered in time, each one is examined to see if it occurs within the current time interval. If it does not, the serial number of the projectile is put into a table at a place based on the proper time interval. The collision is re-evaluated later: events occurring in the cascade between two evaluations often cause them to differ. If the collision occurs within the current time interval, any progeny are placed in the table in the current interval, and the projectile is followed again if it is still moving. Otherwise, an atom is

chosen from the table. For each time interval, the atoms in the table are taken in reverse order to that in which they were entered. All collisions within a time interval are considered simultaneous. The order in which collisions are evaluated depends on the interval used. If it is very large, a particle will be followed to the end of its flight, then the last of its progeny will be followed, and so on. The time interval must be small enough to give a reasonable picture of the time evolution of the cascade. Since most collisions are evaluated twice, time-ordered calculations take up to twice as long as velocity-ordered ones.

2.2. Event and defect classification and time analysis

Collisions are sorted into three groups. *Displacements* are events from which target and projectile both emerge: to be displaced, an atom must receive enough kinetic energy to overcome a binding energy E_b and still retain more than the value E_c at which trajectories are terminated. In *replacement* events, a target emerges, but the projectile stops. The target must overcome only the energy $E_b' < E_b$ to allow for the binding of the stopped projectile. A value is chosen for E_b' so that linear collision sequences (LCSs) in MARLOWE resemble those in dynamical models (see [11] for a discussion). *Focuson* events are a subclass of replacement in which mass is not transported: the target returns to its site after some collisions in its vicinity.

Since the numbers of atoms and sites are conserved in MARLOWE, there are (usually) equal numbers of each. At the end of a cascade, the atoms and sites are arranged in (Frenkel) pairs and sorted into several classes [5]. The arrangement in each cascade is

unique. If an atom originated from its paired site, the pair is *correlated*. If the site in a pair is the nearest in the crystal to the atom, a *close pair* is identified. If the site in a pair is a neighbor of the site nearest the atom, a *near pair* is identified. All other configurations, termed *distant pairs,* are organized by separation. The correlated close and near pairs resemble focuson events and both resemble the sites at which subthreshold encounters occur, while the uncorrelated close and near pairs resemble replacements. It is often appropriate to group the pairs as their morphologies suggest.

The time when a new atom is added to a cascade is part of the data describing its original site. The time when an atom stops is part of its description. At the end of a cascade, such data are used to obtain distributions of the times of the collision events and of the formation of defect pairs, sorted into the classes described above, and from these distributions the number of particles in motion at any time can be obtained.

When stopped cascade atoms are allowed to become targets in later collisions, the times of such *redisplacement* events are recorded separately. No binding energy is associated with this process. The redisplaced atoms are regarded as in motion from the time of their original creation until that of their final termination. No details are retained about their intermediate states.

2.3. Other aspects of the calculation

Collision cascades were generated in static single crystals of Cu and Au; the fcc lattice constant a_0 was 0.3615 and 0.4078 nm, respectively. For each set of initial conditions, 1000 primaries were

launched isotropically from lattice sites with initial kinetic energies up to 10 keV. The Molière potential described the elastic scattering; the screening length a_{12} was 7.38 in Cu and 7.50 pm in Au [1,11]. "Local" inelastic energy losses [3] were based on the LSS theory [12]. The binding energy E_b was 3.50 in Cu and 3.81 eV in Au. In both cases, $E_b' = 0.1\ E_b$ and $E_c = E_b$. For most time-ordered calculations, the time interval was Δt = 1 fs. In this time, a 10 keV Cu atom moves about 0.68 $\underline{a}_0$ and a 10 keV Au atom moves about 0.24 $\underline{a}_0$. Only the first focuson in a sequence was retained, since later ones produce no defects. The calculations were carried out on a Data General Corporation Eclipse MV/10000™ 32-bit computer.

3. Cascade development without redisplacements

When stopped target atoms are not allowed to become involved in later collisions, the only interferences between trajectories that can occur in MARLOWE are those involving target sites: no site emits more than one atom so projectiles can compete for particular sites and the order of the encounters may be significant. Calculations of <N>, the mean number of atoms in motion in a cascade, are shown in Fig. 1 as a function of time for time-ordered cascades initiated in Cu and Au by primaries of 1, 2, and 5 keV. Each curve is bell-shaped with a pronounced tail on the long-time side. The mean slowing-down time of the primaries [8] occurs near the maximum in <N>, but increases with energy more rapidly than does the latter. Cascades in Au develop more slowly and persist longer than do cascades in Cu at the same energy, as is expected from the greater mass of the Au atoms.

The effect of the collision order is examined in Fig. 2 for 10 keV cascades in Cu and Au. The differences between the two schemes are very small in Cu. They are larger in Au, especially beyond the maximum in <N>. In both targets the differences increase with initial kinetic energy. This behavior is consistent with the idea that cascade interferences are more important at high recoil energy in heavy elements. It is doubtful if time-ordered calculations offer much advantage in cases such as these where cascade interferences are minimized by not allowing redisplacements, especially in view of their significantly greater computing-time requirement.

Figure 3 shows the frequencies with which various events occur in 2 keV time-ordered cascades in Cu. Panel (a) shows that a distinct regime may be identified, corresponding to the bulk of the bell-shaped curves in Fig. 1, in which almost all of the displacement events occur. King and Benedek [13] studied the development of collision cascades in Cu at initial energies up to 0.6 keV, using a molecular dynamics code. They identified a collisional regime which lasted for about 200 fs at 0.5 keV. This is in satisfactory agreement with the MARLOWE result, especially considering the different potentials in the two calculations.

The histogram in panel (a) does not show the persistent tail which is present in the other panels. Panel (b) shows the focuson events: most of these occur too early in time to be associated with replacement sequences and must be low-energy recoils ejected by high-energy projectiles. The replacement events, shown in panel (c), begin later than the displacements, since the recoils must slow down to fairly low energies before replacements are probable. At 2 keV, Cu

recoils first produce replacements below about 165 eV. Once replacements begin, they persist, in the form of long LCSs, to very long times, as panels (c) and (d) show. An LCS terminates eventually with an interstitial atom, sometimes followed by a series of focusons. This accounts for the persistent tails in panels (e) and (f). As noted above, sequences of focusons are not followed beyond the first, which weakens the tails in panels (b) and (f).

In view of the small effect of the collision order on <N>, it is not surprising that it has little effect on other cascade properties. Table 1 compares the mean number of distant pairs calculated for velocity-ordered and time-ordered cascades in Cu and in Au at several initial energies. The difference between the velocity-ordered and the time-ordered results is not statistically significant in Cu up to 5 keV; the difference at 10 keV is significant at the 3 σ level. In Au, the corresponding differences are significant at the 4 σ level at 5 keV and at the 9 σ level at 10 keV. In all cases where the difference is sig-nificant, the number of distant pairs calculated in the time-ordered cascades is less than in the velocity-ordered ones.

The greater number of defects in the velocity-ordered calculations can be traced to the fact that the low-energy trajectories are not constructed until those of the faster particles are nearly complete. As a consequence, the low-energy particles encounter more vacant sites than they do in the time-ordered calculations, move slightly farther, and have a greater likelihood of becoming distant pairs, rather than pairs of other types. In time-ordered calculations, the trajectories of low-energy particles are constructed almost from the beginning of the calculation and these trajectories are more likely

to move in undamaged crystal. Thus, it appears to be a general result that less damage will be calculated when the collisions are time-ordered than when they are velocity-ordered.

The effect of varying Δt in the time-ordered calculation is shown in Table 2, which gives the mean number of distant pairs calculated for 1 keV cascades in Au, using several time intervals. It is clear that no significant change occurs for Δt up to 10 fs and this supports the adequacy of the 1 fs value used for most calculations. At energies above 10 keV, it might be necessary to use a smaller Δt.

IV. Cascade development with redisplacements

The subject of cascade nonlinearities can be approached in a limited way in MARLOWE by allowing stopped cascade atoms to become targets in later collisions. Calculations of <N> as a function of time are shown in Fig. 4 for 5 keV time-ordered cascades in Cu and Au, both with and without the redisplacement of stopped cascade atoms. The distribution of redisplacements in time is also shown. Near the maximum in <N> there appear to be more particles moving in cascades with redisplacements, a difference resulting mainly from the fact that redisplaced atoms are not regarded as stopped during their intermediate states. More significantly, beyond ~175 fs in Cu or ~240 fs in Au, <N> is lower with redisplacements than without. This reflects a loss of energy in the redisplacements which is not translated into other forms of motion, particularly long LCSs.

The frequencies with which various events occur is shown in Fig. 5 for 5 keV time-ordered cascades in Au with redisplacements. These histograms are similar to those in Fig. 3 and their inter-

pretation is similar. The distribution of displacements in panel (a) shows a more pronounced tail than in Cu and the tails in the other distributions are somewhat less important. The distribution of redisplacements shows them to occur mainly in the latter part of the displacement phase of cascade development and to disappear before the LCS phase is well-established.

Table 3 compares the mean number of distant pairs calculated at several energies in velocity-ordered and time-ordered cascades with redisplacements. As in Table 1, there are fewer defects in the time-ordered cascades than in the velocity-ordered ones, at least partly because of vacancy effects, as described before. Comparison of Tables 1 and 3 shows a decrease in the number of defects when redisplacements are allowed. More redisplacements occur in time-ordered cascades, so this is further cause of the differences between the two schemes of ordering.

Table 4 shows the effects of the ordering scheme and of the time interval on the mean number of distant pairs produced and also on the number of redisplacements that occur in 1 keV cascades in Au. The velocity-ordered cascades have less than half the redisplacements that the time-ordered cascades do. This comes about because cascade atoms stop from the earliest phase of cascade development when the collisions are time-ordered and are available as potential targets in far more collisions than is the case when the collisions are velocity-ordered. Thus, the increased computational labor in ordering the collisions in time appears justified by the improved modelling.

Redisplacement events mainly involve encounters of slowly moving projectiles with stopped ones. This is shown by several

cascade properties. The maximum energy at which redisplacements occur is only 66 ± 1 eV for 5 keV cascades in Au and the mean kinetic energy of the 79.86 ± 0.69 redisplaced atoms is 4.18 ± 0.06 eV each. In addition, if the value of E_c is increased, the number of redisplacements drops rapidly and nearly vanishes for E_c values above 30 eV or so.

5. Conclusion

The possibility of a BCA model in which the collisions are properly ordered in time has been demonstrated. The results of calculations show that the MARLOWE model contains a nonlinearity associated with the lattice sites (that is, with the conservation of particle number) and that a further nonlinearity involving the collision of cascade atoms with previously stopped atoms can be included. Extension of the model to include encounters between moving atoms may be feasible.

6. Acknowledgement

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7. References

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Table 1
The mean number of distant pairs calculated for displacement cascades without redisplacements using velocity-ordered or time-ordered collisions

E	Cu		Au	
(keV)	V	$\Delta t = 1$ fs	V	$\Delta t = 1$ fs
0.5	8.82±0.06	8.84±0.06	5.38±0.06	5.43±0.04
1	17.98±0.09	18.03±0.08	9.49±0.10	9.39±0.08
2	34.73±0.12	34.69±0.12	18.95±0.14	18.83±0.14
5	81.81±0.19	81.49±0.18	50.22±0.21	48.89±0.20
10	156.61±0.26	155.45±0.26	99.26±0.29	95.52±0.27

Table 2
Effect of the time interval on the mean number of distant pairs in 1 keV cascades in Au without redisplacements

Time Interval (fs)	Mean Number of Distant Pairs
0.1	9.35±0.11
1	9.39±0.08
10	9.40±0.10
100	9.15±0.10

Table 3
The mean number of distant pairs calculated for displacement cascades with redisplacements using velocity-ordered or time-ordered collisions.

E	Cu		Au	
(keV)	V	$\Delta t = 1$ fs	V	$\Delta t = 1$ fs
0.5	8.77±0.06	8.76±0.06	5.27±0.05	5.18±0.05
1	17.82±0.09	17.73±0.08	9.26±0.10	8.98±0.10
2	34.38±0.12	34.10±0.11	18.47±0.14	17.87±0.13
5	80.78±0.19	79.57±0.17	48.11±0.19	45.07±0.18

Table 4
Effect of the time interval on the mean number of distant pairs in 1 keV cascades in Au with redisplacements

Time Interval (fs)	Mean Number of Distant Pairs	Mean Number of Redisplacements
0.1	8.91±0.10	7.69±0.20
1	8.98±0.10	7.67±0.18
10	8.98±0.10	7.18±0.17
100	8.84±0.10	7.45±0.17
V	9.26±0.11	3.36±0.09

Figure Titles

Fig. 1. Time-ordered displacement cascades in Cu and Au: the mean number of projectiles in motion as a function of time.

Fig. 2. 10 keV displacement cascades in Cu and Au: the mean number of projectiles in motion as a function of time, comparing calculations with velocity-ordered collisions with those for time-ordered collisions.

Fig. 3. 2 keV displacement cascades in Cu: the frequencies of occurrence of different types of events, as functions of time. The hatched boxes at the right of each panel show the particles omitted from each histogram. The mean number of each kind of event per cascade is shown in each panel.

Fig. 4. 5 keV displacement cascades in Cu and Au: the mean number of projectiles in motion as a function of time, comparing time-ordered calculations with and without redisplacements.

Fig. 5. 5 keV displacement cascades in Au: the frequencies of occurrence of different types of events, as functions of time. The hatched boxes at the right of each panel show the particles omitted from each histogram. The mean number of each kind of event per cascade is shown in each panel.

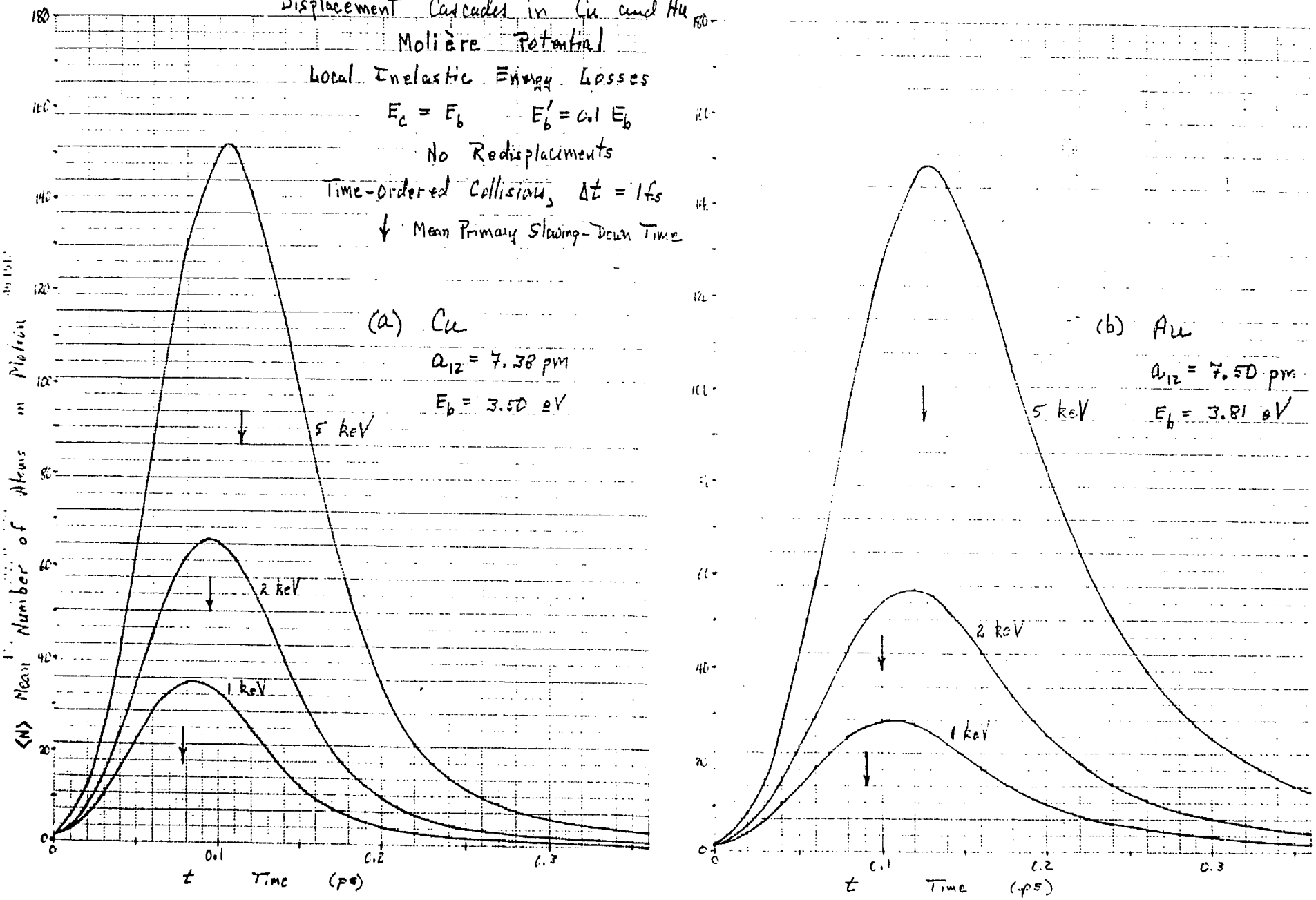

Displacement Cascades in Cu and Au
Molière Potential
Local Inelastic Energy Losses
$E_c = E_b$ $E_b' = 0.1\ E_b$
No Redisplacements
Time-ordered Collisions, $\Delta t = 1$ fs
↓ Mean Primary Slowing-Down Time
(a) Cu
$a_{12} = 7.38$ pm
$E_b = 3.50$ eV
5 keV
2 keV
1 keV
$\langle N \rangle$ Mean Number of Atoms in Motion
t Time (ps)
(b) Au
$a_{12} = 7.50$ pm
$E_b = 3.81$ eV
5 keV
2 keV
1 keV
t Time (ps)

FIG. 1

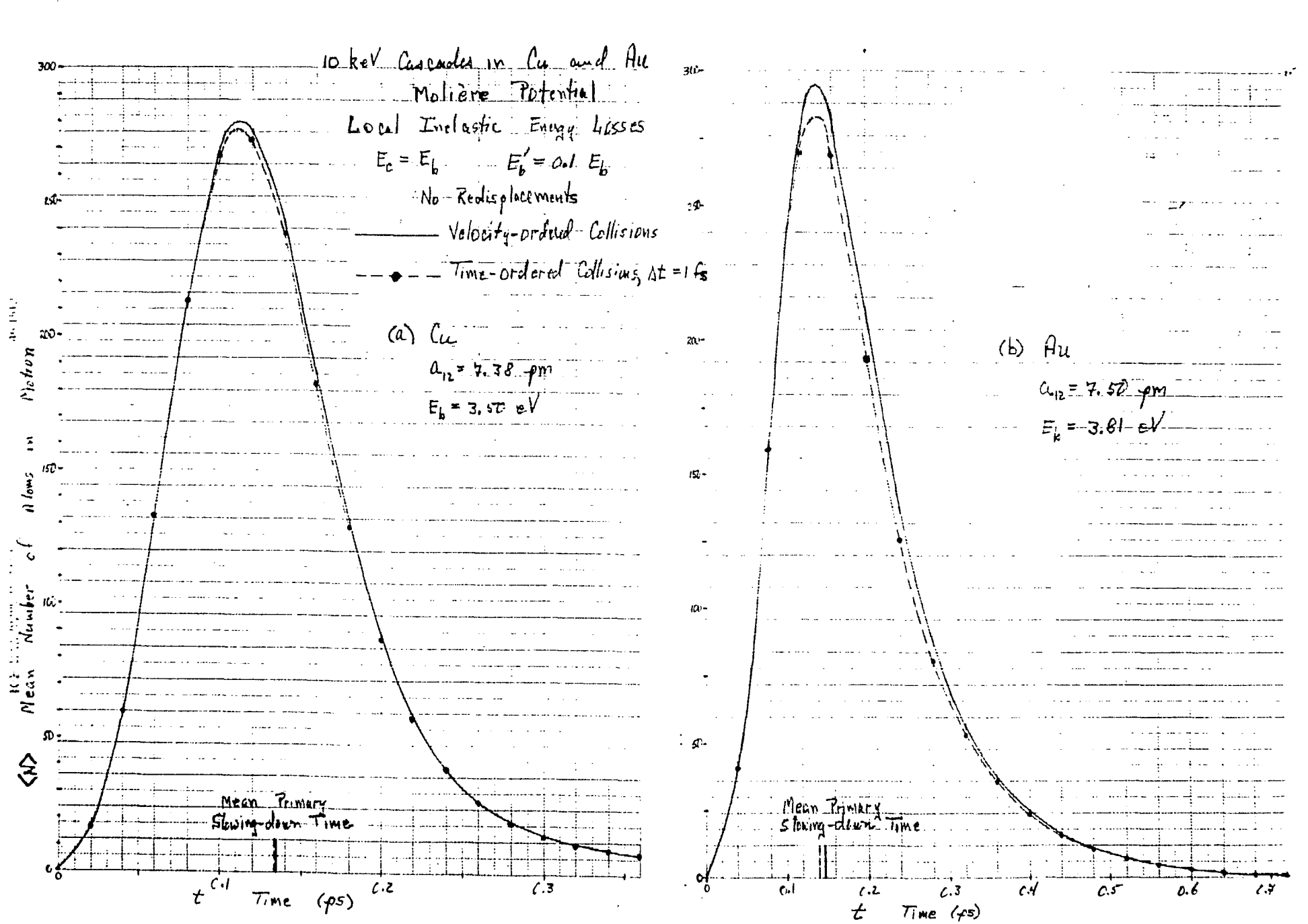

10 keV Cascades in Cu and Au
Molière Potential
Local Inelastic Energy Losses
$E_c = E_b$ $E_b' = 0.1\ E_b$
No Redisplacements
Velocity-ordered Collisions
Time-ordered Collisions, $\Delta t = 1$ fs
(a) Cu
$a_{12} = 7.38$ pm
$E_b = 3.57$ eV
(b) Au
$a_{12} = 7.52$ pm
$E_b = 3.81$ eV
Mean Primary Slowing-down Time
Mean Primary Slowing-down Time
$\langle N \rangle$ Mean Number of Collisions per Motion
t Time (ps)
t Time (ps)

Fig. 2

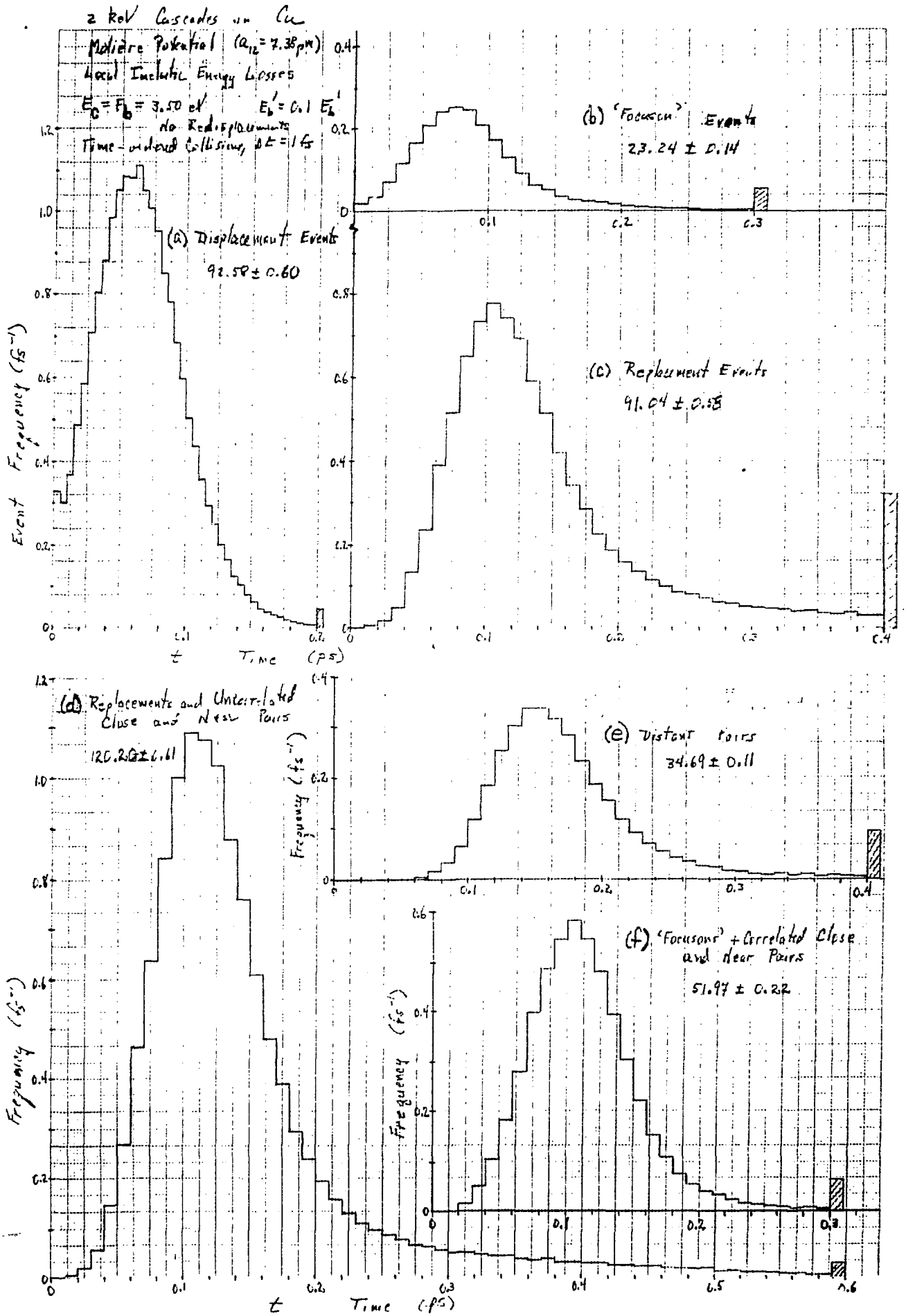

2 keV Cascades in Cu
Moliere Potential (a_{12} = 7.38 pm)
Local Inelastic Energy Losses
$E_0 = E_b = 3.50$ eV $E_b' = 0.1\ E_b$
No Replacements
Time-ordered Collisions, Δt = 1 fs
(a) Displacement Events
92.58 ± 0.60
(b) 'Focuson' Events
23.24 ± 0.14
(c) Replacement Events
91.04 ± 0.58
(d) Replacements and Uncorrelated Close and Near Pairs
120.20 ± 0.61
(e) Distant Pairs
34.69 ± 0.11
(f) 'Focusons' + Correlated Close and Near Pairs
51.97 ± 0.22
Event Frequency (fs⁻¹)
Frequency (fs⁻¹)
t Time (ps)

Fig. 2

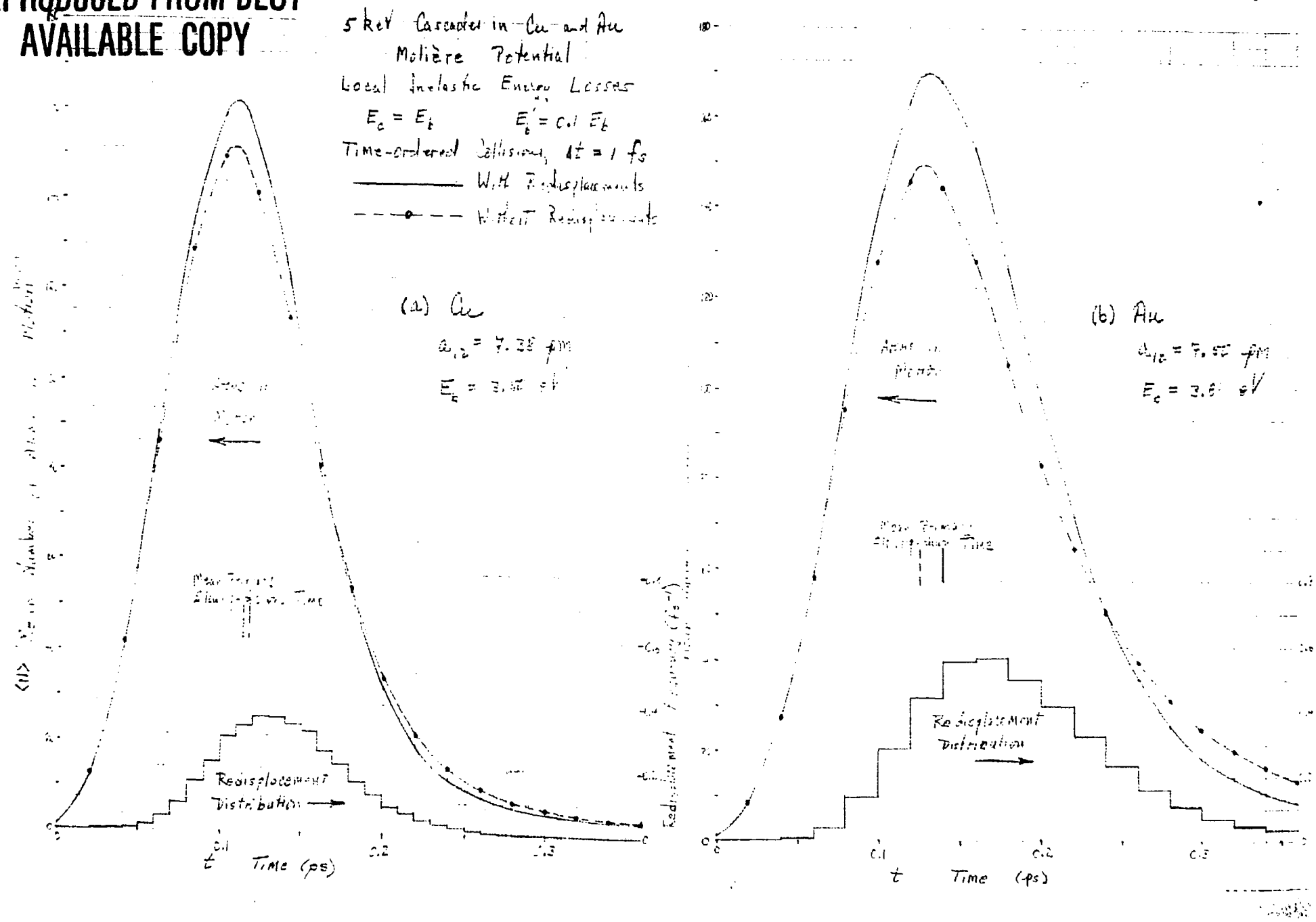
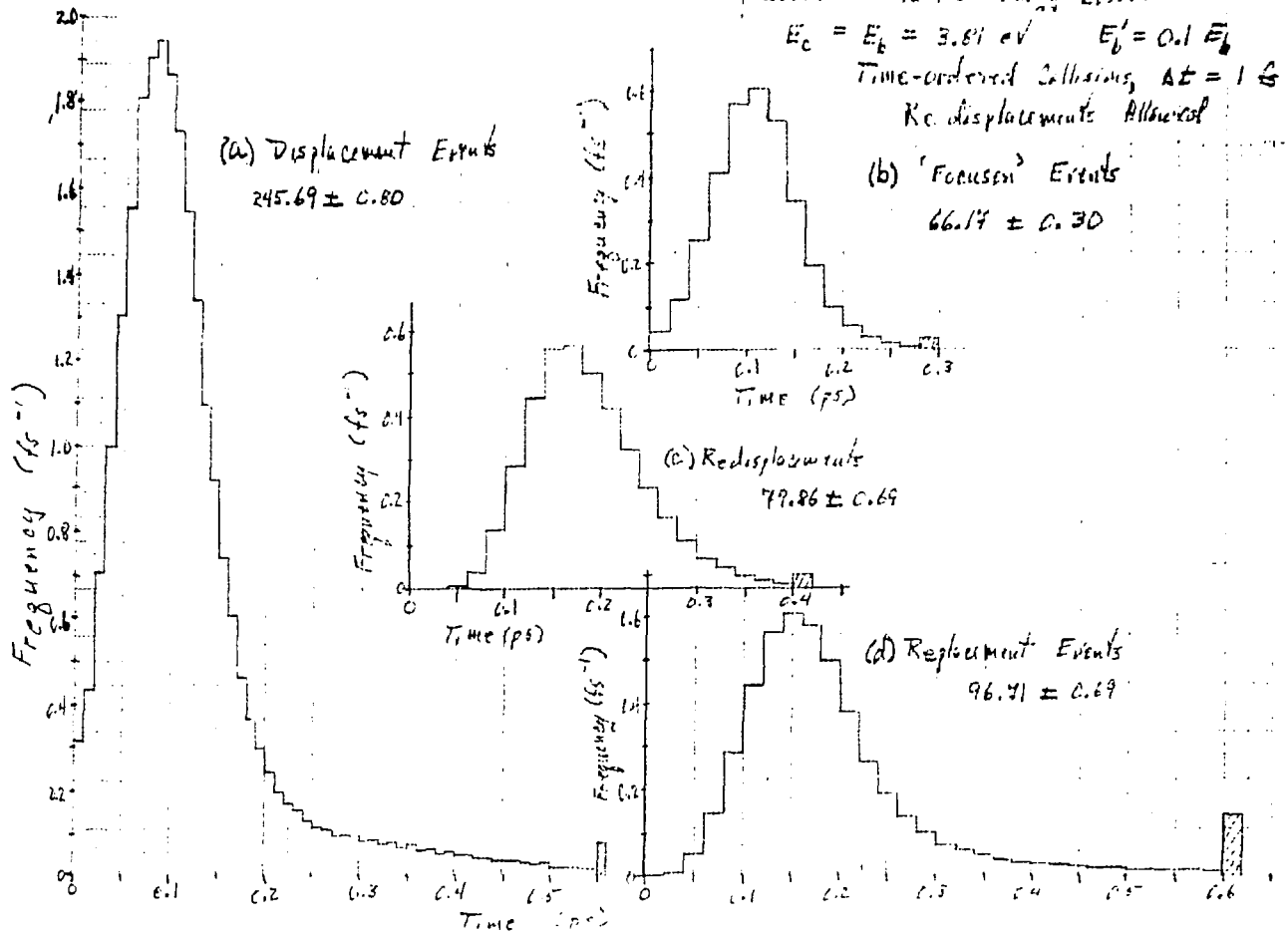


Fig. 4

5 keV Cascades in Ni
Molière Potential (a_{12} = 7.20 pm)
Local Inelastic Energy Losses
$E_c = E_b = 3.81$ eV $E_b' = 0.1\,E_b$
Time-ordered Collisions, $\Delta t = 1$ fs
Re-displacements Allowed



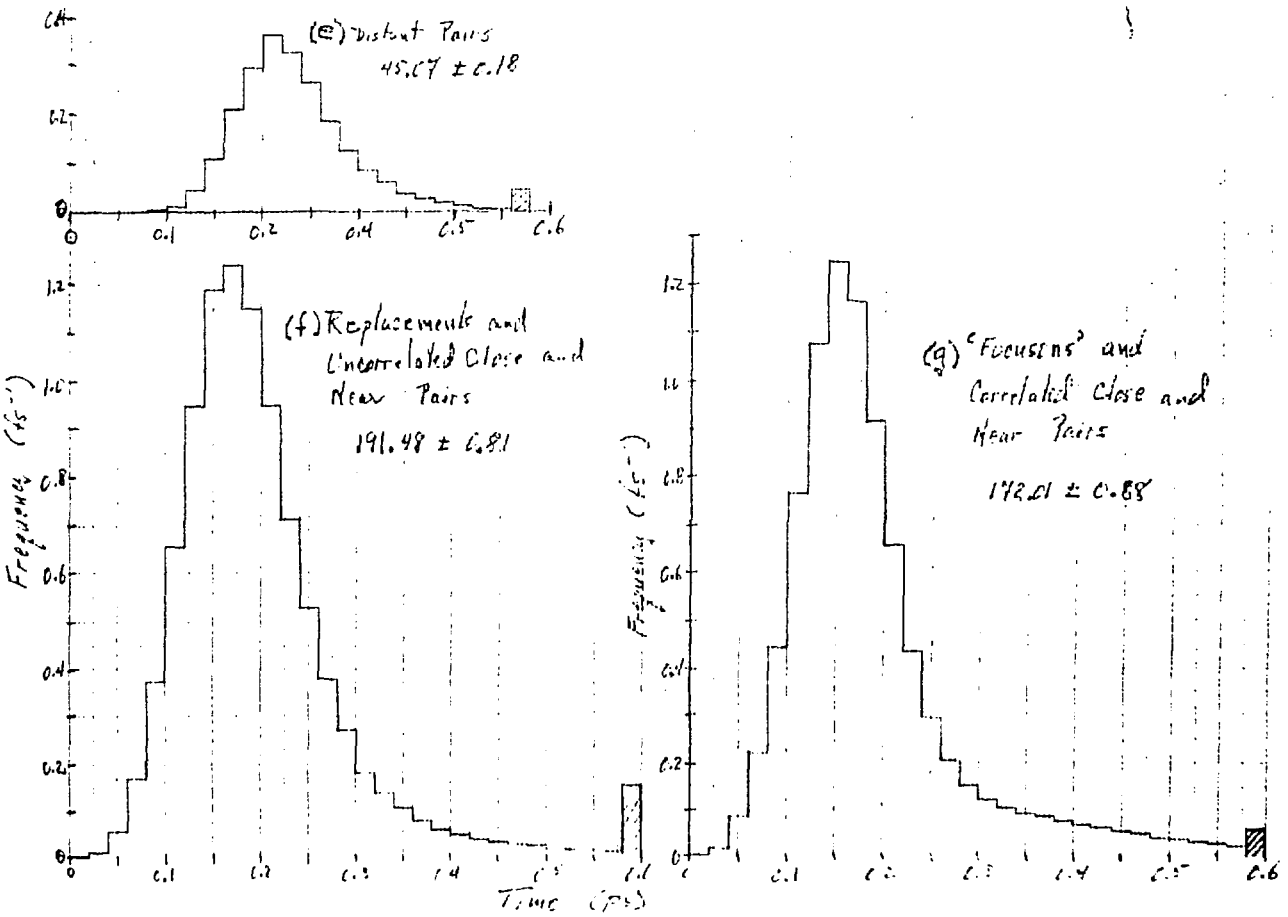


Fig. 5