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IN-SITU STUDIES OF CASCADE DEFECT FORMATION AT T < 10 K†

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This work is being submitted to the Seventh International Conference on High Voltage Electron Microscopy, August 16-19, 1983, Berkeley, CA.

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ABSTRACT

Thin films of Ag and Au were ion-irradiated at temperature (T) < 10 K and simultaneously observed using the Argonne National Laboratory high-voltage electron microscope (HVEM) ion-beam interface system. Displacement cascades were produced by Kr^+ ion bombardment in the energy range 20-140 keV. For Ag, 20% of the 20-keV cascades and 100% of the 100-keV cascades produced observable contrast. Analysis of the observed dynamical black-white (BW)-contrast indicates that the clusters are of vacancy type. Subcascades were observed in the high-energy irradiations. The present results demonstrate that, during the evolution of displacement cascades in Ag and Au, extensive athermal rearrangement of vacancies occurs which gives rise to clustering and "collapse" to dislocation loops.

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INTRODUCTION

The microstructural and microchemical evolution of materials under heavy ion or fast neutron irradiation depends strongly on the spatial distribution of defects in individual displacement cascades. The classical cascade model is that of a vacancy rich depleted zone surrounded by interstitials [1]. However, evidence exists that this simiple picture has to be considerably modified. Previous room temperature transmission electron microscopy (TEM) observations of displacement cascades in Ag, Au and a number of other metals have shown considerable clustering of vacancies and agglomeration to dislocation loops [2,3]. The clustering either occurs during the time evolution of the cascade or is due to thermally stimulated processes. Electrical resistivity measurements at liquid helium temperature suggest a significant amount of athermally stimulated recombination of point defects due to atomic motion during the thermal spike phase of the cascade [4]. The existence of such spike effects was also shown in TEM observation of surface craters in Au bombarded with Bi and Bi_2 ions at room temperature [5]. However, direct TEM observation of cascade formation at low temperature has

not been carried out to confirm the existence of such spike effects.

In the present work, Ag and Au thin films were irradiated in-situ in the Argonne National Laboratory HVEM ion-beam interface system at T < 10 K. The existence of athermally stimulated rearrangement and collapse of the vacancy rich core of the cascade was confirmed by direct observation at low temperature.

EXPERIMENTAL

Thin film (~ 1000 Å thick) specimens of Ag and Au were prepared by vacuum evaporation and subsequent annealing. These films were irradiated with Kr^+ ions at T < 10 K in the HVEM using the ion-beam interface system at Argonne National Laboratory [6] and the single tilt He temperature specimen stage [7]. Ion energies were 20 keV and 100 keV for Ag and 140 keV for Au. The irradiations were interrupted several times and TEM observations were carried out without warming the specimen. The ion dose was measured using a transmission Faraday cup which intercepts an annular fraction of the beam and is positioned at the end of the ion-beam line inside the HVEM column, just above the specimen stage (the ion beam axis was inclined 33° with respect to the electron beam axis). Ion beam rastering in two orthogonal directions perpendicular to the beam axis was employed to ensure that the integrated beam current density was homogeneous over the 3 mm diameter entrance aperture of the cup. The highest electron energy used for TEM observation was 500 keV which is well below the displacement threshold in both Ag and Au. At the electron current densities (~ 10^{18} e/cm^2/sec) used, the specimen temperature rise due to electron beam heating was calculated to be negligible [8]. Micrographs were taken at specimen orientations near [111] (Ag) and [100] (Au) under kinematical bright field conditions and also dynamical bright and dark field conditions. The defect type was determined from the direction of $\vec{\ell}$, the vector from the black lobe to the white lobe of the BW-contrast under dynamical conditions, relative to the diffraction vector $\vec{g}$ [9]. The defect yield, the fraction of displacement cascades which form visible defect clusters, was evaluated by measuring the areal density of defect clusters or subcascade structures on micrographs and comparing it with the measured ion dose. TEM observations of the same areas were also performed after warming the irradiated specimen to room temperature.

RESULTS

Figure 1 shows the BW-contrast of cascade defect clusters in Ag irradiated with 20 keV Kr^+ ions at T < 10 K. All the BW-contrasts observed had $\vec{g} \cdot \vec{\ell} > 0$. The calculated mean penetration depth (using the TRIM code [10]) for 20 keV Kr^+ ions in the Ag specimens was 65 Å, well within the first layer of the depth oscillation of BW-contrast (the approximate thickness of the first layer is $0.3\,\xi_g$ = 164Å, when imaged with $\vec{g} = 2\bar{2}0$ at an electron energy of 400 keV [11]). The clusters were therefore identified as being of vacancy type. Three different $\vec{\ell}$ vectors were found. One was parallel to $\vec{g}$, others were close to the projection of the $[1\bar{1}1]$, or $[\bar{1}11]$ direction onto the (111) plane. Clusters which have $\vec{\ell}$ parallel to $\vec{g}$ may have a spherically symmetrical strain field and may be small stacking fault tetrahedra [12,13] or uncollapsed vacancy clusters [14]. Other clusters are thought to be Frank dislocation loops with Burgers vector $\vec{b} = \frac{a}{3}\langle 111\rangle$. Subcascade formation, i.e. the splitting of a cascade into multiple depleted

zones, was observed for 100 keV cascades in Ag (Fig. 2. (a), (b)) and 140 keV cascades in Au (Fig. 3). The yield of visible cascades in Ag was 0.2 for 20 keV irradiation and 1.0 for 100 keV irradiation (closely spaced subcascades were counted as one). The defect density increased linearly with ion dose. Fig. 2 illustrates the development of the defect structure with dose. Subcascade structures during in-situ observation (Fig. 2. (a), (b)) always appeared simultaneously, indicating that they are associated with individual ion impacts. Fig. 4 shows 100 keV cascades in Ag after warming to room temperature. No significant changes in the defect density were observed when warming the specimen to room temperature.

DISCUSSION

The defect yield values can be compared with previous data on self-ion bombardment of Ag at room temperature [14]. For 100 keV both the room temperature and low temperature results indicate a cascade yield of 1. For 20 keV irradiation the present yield (0.2) was slightly lower than that found in room temperature self-ion irradiation (0.35 Ref. [14]). This is probably not due to an ambient temperature effect, since a lower yield is expected due to the lower energy density of Kr^+ ion cascades compared to self-ion cascades. The existence of defects which were out of contrast under kinematical bright field conditions used for defect density measurements [12] could also give lower yield values, although the comparison with micrographs of the same areas taken under dynamical conditions indicated that only a small fraction of the defects was out of contrast.

The present results demonstrate that in Ag and Au vacancies in an individual cascade are clustered and collapse into Frank dislocation loops even at low temperature where the thermal migration of point defects is suppressed. A possible explanation for this very effective rearrangement of vacancies is given by thermal spike effects during the evolution of displacement cascades. After the cascade establishes its branching sequence of collisions producing many Frenkel pairs (typical time $\sim 10^{-13}$ s [15]), the kinetic energy of knocked-on atoms dissipates into the surrounding lattice. In the early stages (lifetime $\sim 10^{-12}$ s [15]), most of the energy is shared with the atoms in the immediate vicinity. Therefore the kinetic energy is still contained within a small volume comparable to the volume of damage straggling. During this period the energy density, i.e. average energy per atom in the volume, is so high that rearrangements of interstitial atoms and vacancies can take place by diffusional or quasi-diffusional jumps. Since the lifetime of this excited state is only on the order of a few tens of lattice vibration periods, it seems apparent that this atomic agitation is also accompanied by collective interactions which then lead to the observed extensive clustering of vacancies within the depleted zone and the collapse into a Frank dislocation loop as a stable configuration.

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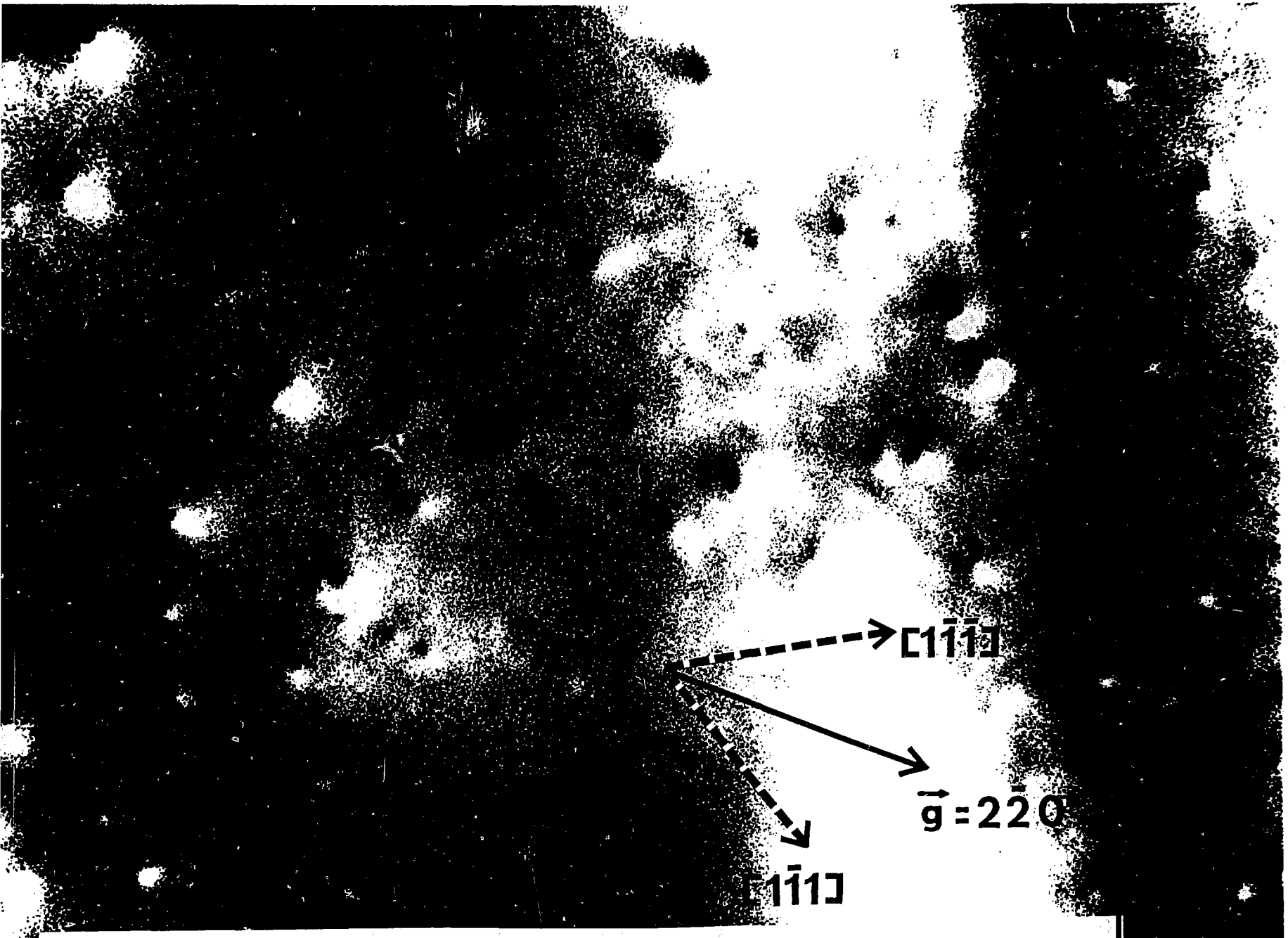

Fig. 1. Black-white contrast of cascade vacancy clusters in Ag irradiated with 20 keV Kr^+ ions at T < 10 K, dose = 1.2 x 10^{11} ions/cm^2, microscopy at T < 10 K with 400 keV electrons, dynamical dark

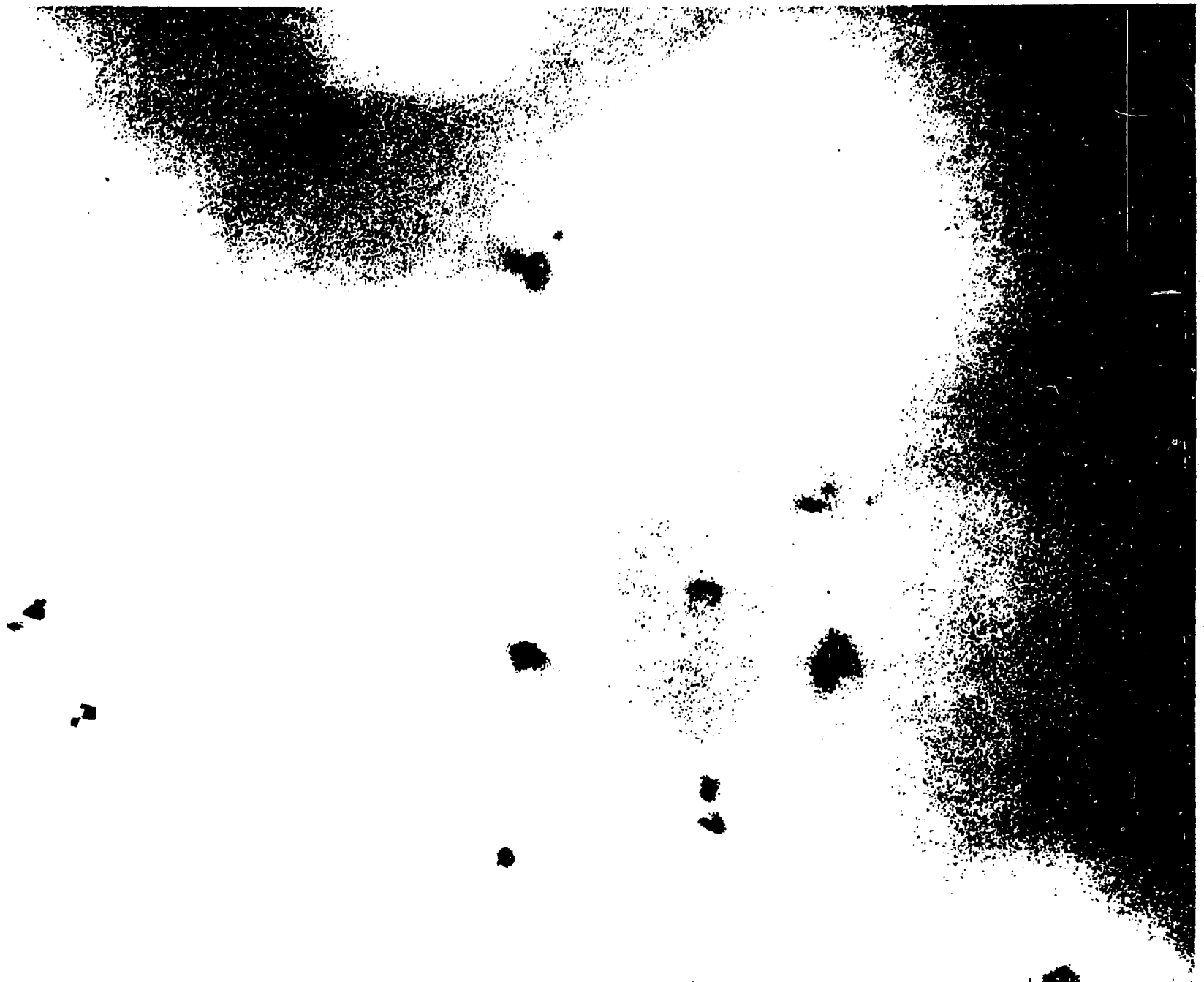

Fig. 2. Cascade vacancy clusters in Ag irradiated with Kr^+ ions at T < 10 K, microscopy at T < 10 K, kinematical bright field.
100 keV Kr^+ → Ag: (a) 2.3 x 10^9 ions/cm^2, (b) 5.6 x 10^9 ions/cm^2;
20 keV Kr^+ → Ag: (c) 5.5 x 10^{10} ions/cm^2, (d) 1.2 x 10^{11} ions/cm^2.


500A

(c)


500Å
(d)

Fig. 3. Cascade defect clusters in Au irradiated with 140 keV Kr^+ ions at T < 10 K, microscopy at T < 10 K, kinematical bright field.

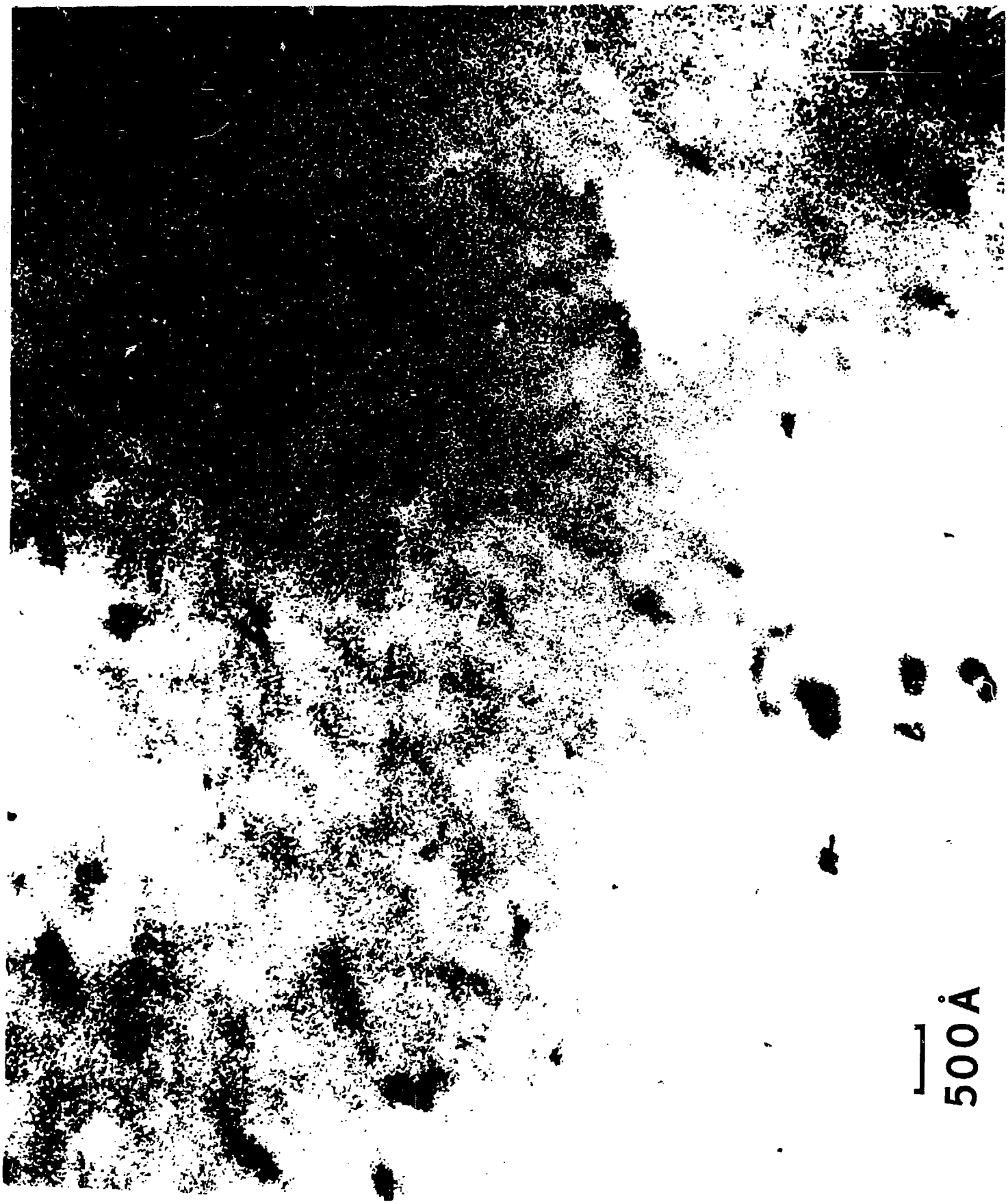

Fig. 4. Cascade vacancy clusters in Ag warmed to room temperature after 100 keV Kr^+ ion irradiation at T < 10 K, kinematical bright field, dose = 1.2×10^{10} ions/cm^2.