BAP-25

ION-BEAM-ENHANCED ADHESION IN THE ELECTRONIC STOPPING REGION*

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ABSTRACT

We report here the first use of ion beams in the electronic stopping region to improve the adhesion of insulators to other materials. In particular, we have dramatically improved the bonding of Au films to teflon, ferrite, and SiO_2 by bombarding them with He and Cl, respectively. Improvements in bonding have also been observed for Au on glass, Au and Cu on sapphire, and Si_3N_4 on Si. The mechanism is apparently associated with sputtering and track-forming processes occurring in the electronic stopping region. Numerous applications are discussed.

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ABSTRACT

We report here the first use of ion beams in the electronic stopping region to improve the adhesion of insulators to other materials. In particular, we have dramatically improved the bonding of Au films to teflon, ferrite, and SiO_2 by bombarding them with He and Cl, respectively. Improvements in bonding have also been observed for Au on glass, Au and Cu on sapphire, and Si_3N_4 on Si. The mechanism is apparently associated with sputtering and track-forming processes occurring in the electronic stopping region. Numerous applications are discussed.

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Ion-beam-induced atomic mixing is a well known technique for improving the bonding at an interface between dissimilar materials. Until now the ion beams used for mixing have had energies in the nuclear stopping region. In recent years an extraordinarily powerful sputtering mechanism has been found to operate in the electronic stopping region in a large class of insulators$^{1-4}$). Consequently, we suspected that ion bombardment in the electronic stopping region could be used to improve the bonding between an insulator and any other solid. In this letter we report on several systems that exhibit improved bonding after very high energy bombardment: Au on teflon, Au on SiO_2, Au on ferrite, Au on soda-lime glass, Au on Al_2O_3, Cu on Al_2O_3, and Si_3N_4 on Si.

The experimental techniques required to demonstrate the bonding are simple. Commercial grade teflon, ferrite, fused quartz, sapphire, and soda-lime glass were cleaned with trichloroethylene, nitric acid and methanol before being loaded into a diffusion-pumped evaporator. We evaporated 200 Å-500 Å of Au or Cu onto the samples in a vacuum of 1×10^{-6} Torr. The Si_3N_4 films on Si were formed by sputter deposition in an RF discharge sputtering chamber. We irradiated the films with 1 MeV He, 2 MeV H, 5 MeV F and 20 MeV Cl beams from the ONR-CIT tandem accelerator. The beam spot was in most cases defocussed to a horizontal stripe 1 cm long and 0.1 cm wide. A beam sweeper was not available, so the beam dose in each spot was uniform to within a factor of two only. This did not prove to be a serious problem. After the irradiations we tested the adhesion of the films with the "Scotch Tape test": a piece of tape firmly pressed onto the film was slowly pulled off by hand. The effect obtained from the high energy bombardments is so dramatic that more quantitative tests of adhesion were not necessary.

Enhanced bonding has been easiest to produce with Au on teflon. A fluence of $3\text{-}4 \times 10^{13}/cm^2$ He at 1 MeV produces a strong bond. In fig. 1

we show a photograph of a 500 Å Au film on teflon bombarded with 1 MeV alpha particles at fluences of $2 \times 10^{13}/cm^2$, $4 \times 10^{13}/cm^2$ and $8 \times 10^{13}/cm^2$. The beam currents were about 150 nA/cm^2 (+1 charge state), which allowed irradiation times of 2 min or less. The power delivered by the beam was approximately 0.15 $watt/cm^2$. The tape easily pulled the Au from the unirradiated areas. Notice that the Au did not adhere to the central region of the highest fluence spot. Our non-uniform beam tended to be more intense in the center. The teflon cannot withstand fluences that are too high; doses above $5 \times 10^{13}/cm^2$ do not produce improved adhesion at 1 MeV incident energy and 150 nA/cm^2 incident beam current. A fluence of $1 \times 10^{13}/cm^2$ did not produce enhanced bonding when tested immediately after the irradiation, but when tested five days later it produced bonding comparable to the slightly higher doses. Why the apparent improvement appears after aging in air at room temperature is not known; the result may be produced by variations in the adhesion of the tape to the Au film. 2 MeV protons produced excellent results at a dose of about $5 \times 10^{14}/cm^2$, which is almost ten times greater than that required for 1 MeV alphas. A lower proton energy probably would have allowed us to decrease the fluence. F beams at 5 MeV were found to improve the bonding at fluences above $3 \times 10^{12}/cm^2$.

We can enhance the bonding of Au to SiO_2 by using a heavier particle, Cl. This beam was chosen because it is the heaviest one easily available from our accelerator; Ar would have worked just as well. Cl at 20 MeV dramatically improves the bonding with a fluence of $1 \times 10^{15}/cm^2$ (fig. 2). A fluence of $5 \times 10^{14}/cm^2$ produces an effect that is just barely observable with our tape test. Aging the irradiated sample for two weeks in air at room temperature substantially improved the tape test result for $1 \times 10^{15}/cm^2$. Fluorine irradiation had no influence on this combination at doses up to

fig

$1 \times 10^{15}/cm^2$. Thus, the bonding exhibits a threshold not only in the fluence but also in the atomic number of the incident particle. This behavior is similar to that found with registration thresholds in nuclear track production, a phenomenon which is probably related to our bonding mechanism. We also evaporated Au onto a 2000 Å SiO_2 film on a Si substrate. The results from Cl bombardment of this film on a film were identical to those from the bulk SiO_2 samples.

Au on a nickel-zinc ferrite supplied to us by IBM proved to be the most well behaved system that we studied. 5 MeV F at $2 \times 10^{15}/cm^2$ and 20 MeV Cl at $3 \times 10^{13}/cm^2$ produced a strong bond. For both incident beams the adhesion was improved over a wide range of incident fluences. The results from one of our tape tests can be seen in fig. 3.

fig.3

Our tests of Au on glass and of Si_3N_4 on Si produced enhancements that were not as dramatic as on the previous systems. In both cases we used Cl beams. 20 MeV Cl at $1 \times 10^{15}/cm^2$ just barely produced improved bonding onto the glass substrate only after the irradiated spot had aged in air at room temperature for two weeks. Some of our Si_3N_4 films adhered to Si quite well without any help from our ion beams. One batch that did not adhere well, however, was bonded to the Si strongly enough to pass the tape test using Cl at 20 MeV and 5 MeV with a fluence of $1 \times 10^{15}/cm^2$. Very high doses of 20 MeV Cl improved the bonding of Au and Cu to sapphire. About $1 \times 10^{16}/cm^2$ was needed for the Au to pass the tape test, while $3 \times 10^{15}/cm^2$ was required for the Cu. For sapphire a heavier incident particle is clearly preferable.

The mechanism that produces the bonding is not understood, though it is apparently associated with the sputtering mechanism that operates in this energy range. What we know about the sputtering process can give us some clues about the enhanced adhesion. Enhanced bonding should appear in the electronic stopping region only if one or both of the media are electrical insulators, and it should show the greatest effect when the incident ion is near the peak of the electronic stopping power. The moving atoms in the disrupted medium have energies that are much lower than that in the collision cascades produced by low energy ions. If there is mixing at the interface, the mixed layer should not be very thick. Analyses of the sputtered atoms suggest that the [illegible] produced by the incident particle can be characterized by a temperature — typically a few thousand degrees Kelvin[5]. Perhaps the improved bond is a sort of spot weld.

This process lends itself to important but simple applications. There are many situations in which it is necessary to bond a good conductor to an insulator: printed circuit boards, integrated circuits, mirrors, ferrite heads for tape and disk drives, etc. Since many protective coatings such as paints are insulators, the properties of protected surfaces could be improved after the coating has been applied. This mechanism could also be used to bond insulators to insulators for applications such as coated optics. Metallization of polymers is probably the easiest application, since high energy He beams can be produced with a modest apparatus. A one milliamp alpha beam could bond metals to teflon at a rate of over 150 cm^2/sec. Nuclear reactions and alpha emitters could also be used to perform the bombardments. For instance, in a nuclear reactor the $^{10}B(n,\alpha)^{7}Li$, $^{3}He(n,p)T$, $^{6}Li(n,\alpha)T$, and $^{235}U(n,f)$ reactions could be used to process metallized polymers with very large surface areas. Since neutrons can penetrate deep into a solid, the process is not limited to bonding thin films to substrates. In addition, high energy ion beams have advantages over low energy ion-induced mixing. The beam particles travel far beyond the bonded interface so they do not contaminate it. Furthermore, a high energy ion beam would not sputter away the metal film as a low energy beam would. In many ways this high energy mechanism could be more convenient to use than low energy ion techniques.

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FIGURE CAPTIONS

Fig. 1. A 500 Å Au film on teflon bombarded with a 1 MeV ^{4}He beam at fluences of $2 \times 10^{13}/cm^2$, $2 \times 10^{13}/cm^2$, $4 \times 10^{13}/cm^2$ and $8 \times 10^{13}/cm^2$ (from the bottom to the top). "Scotch Tape test" has been done only on the left-hand of the sample and the upper blank was unevaporated area.

Fig. 2. A 500 Å Au film on SiO_2 bombarded with a 20 MeV Cl beam at fluence of $1 \times 10^{15}/cm^2$. "Scotch Tape test" has been done twice on the right-hand of the sample and a substantial improvement can be seen on the second test after aging the irradiated sample for two weeks in air at room temperature.

Fig. 3. A 500 Å Au film on nickel-zinc ferrite bombarded with a 20 MeV Cl beam at fluences of $3 \times 10^{13}/cm^2$, $1 \times 10^{14}/cm^2$ and $3 \times 10^{14}/cm^2$ (from the top to the bottom). "Scotch Tape test" has been done only on the right-hand of the sample and the upper blank was unevaporated area.

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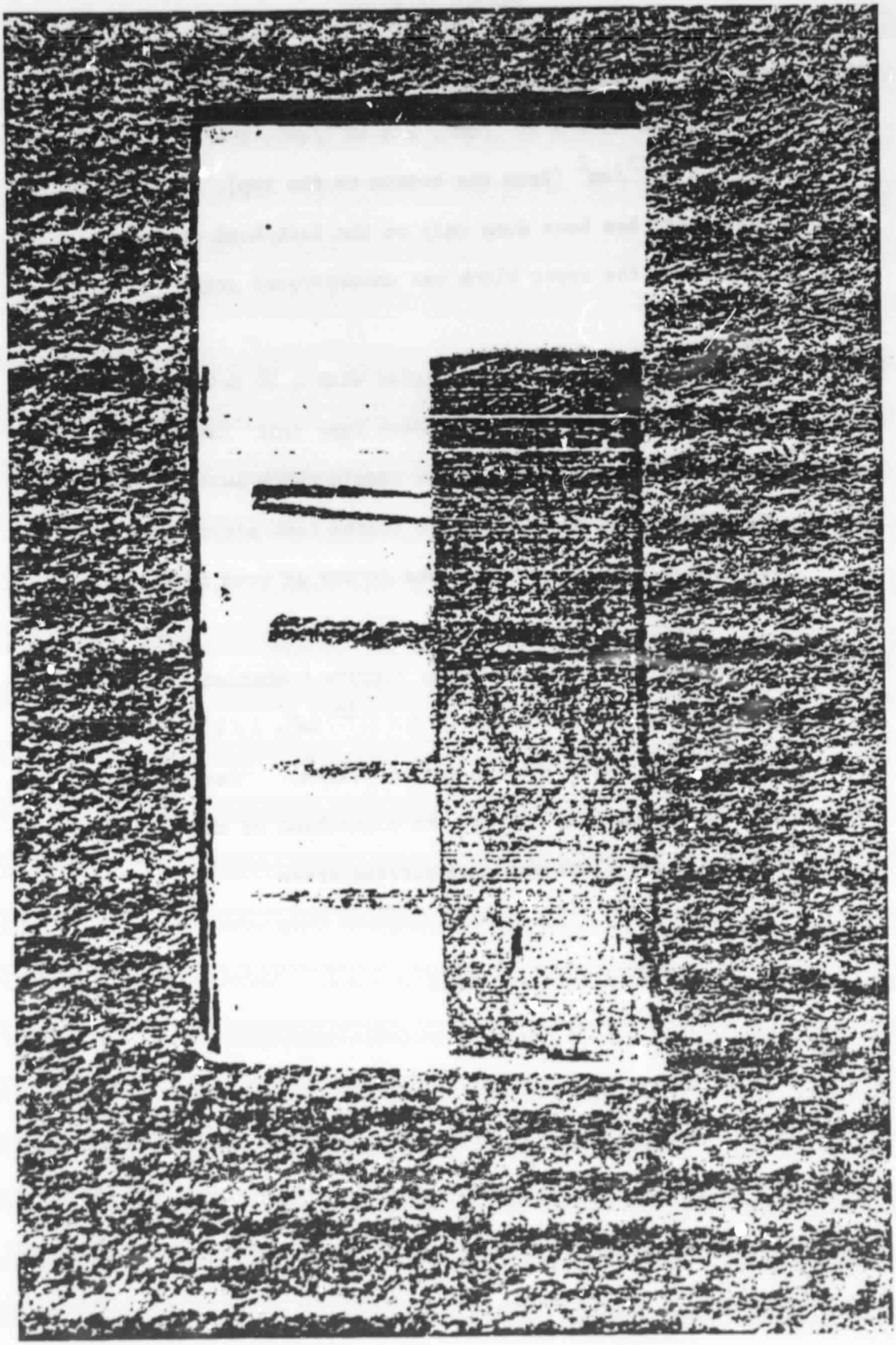

Fig. 1

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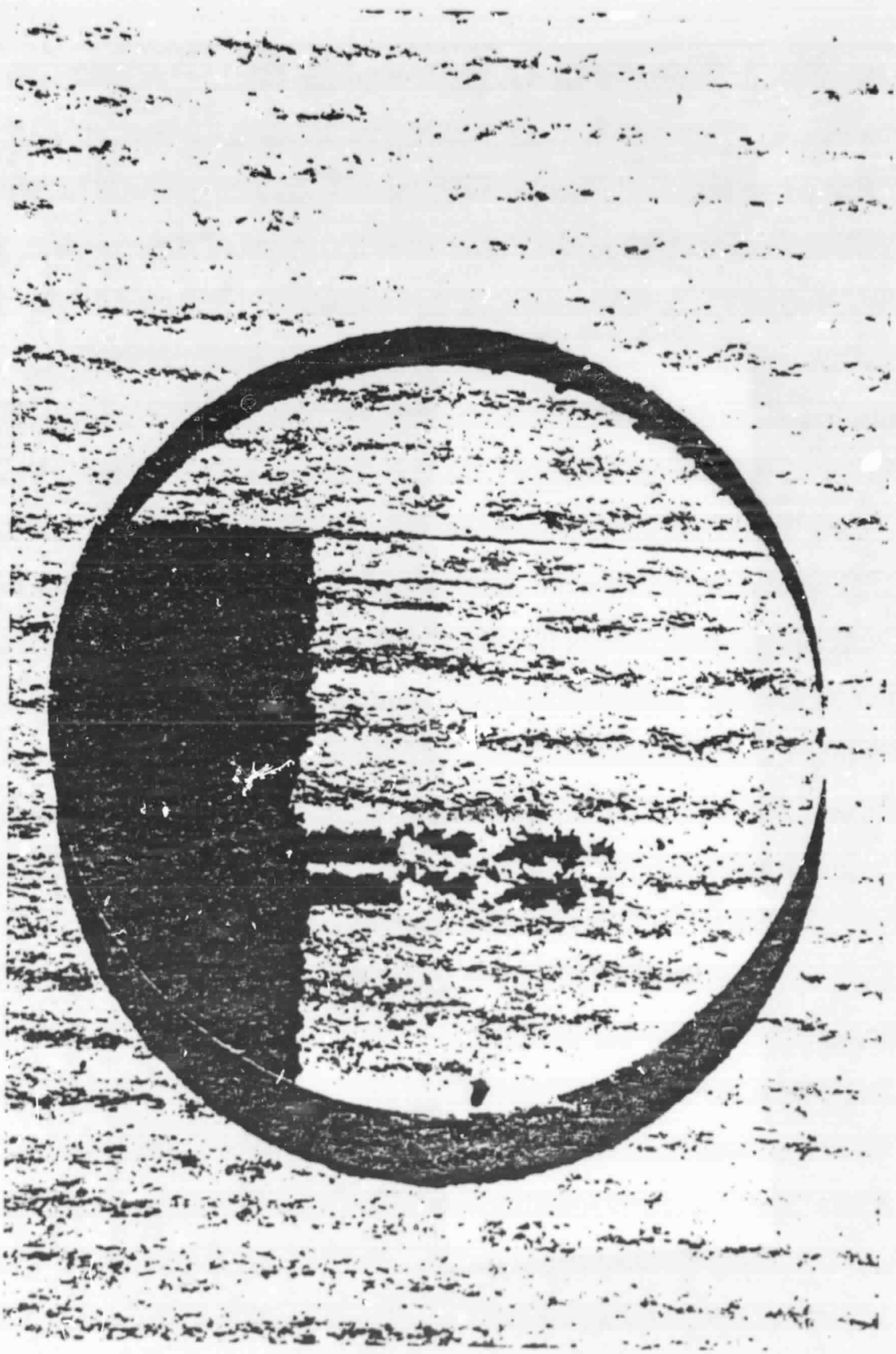

Fig. 2

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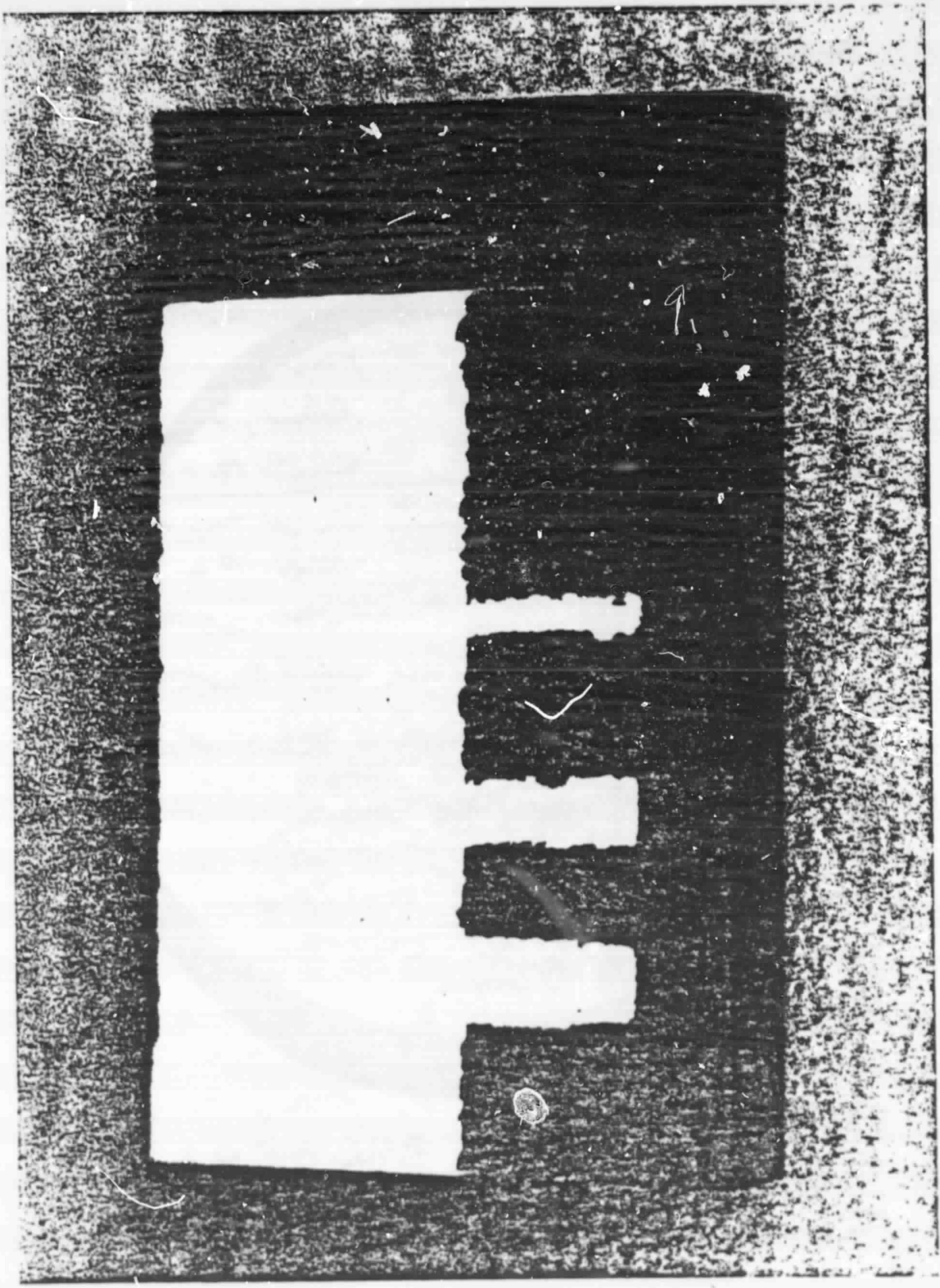

Fig. 3