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ARTIFICIALLY-INDUCED THERMOLUMINESCENCE GRADIENTS IN STONY METEORITES

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Several meteorites show variations in natural thermoluminescence intensity with depth. To investigate this, the gamma-ray induced thermoluminescence of the Plainview, Ucera, and Lost City meteorites has been analyzed. The results show that the thermoluminescence gradients in the meteorites are related to the concentration of electron traps, probably as a result of some of them being produced by cosmic ray bombardment.

INTRODUCTION

Observations of the variation of the natural thermoluminescence (TL) intensity throughout the Saint Séverin, Ucera, Estacado and Allende meteorites have been made by Lalou *et al.* (1970a), Vaz (1971a), Sears (1975a) and Sears and Mills (1974a), respectively. They found a considerable depth dependence in the natural TL intensity of these meteorites which may be interpreted as being due to cosmic-ray bombardment of the meteoroids or to atmospheric heating during the flight of the meteorites. Houtermans and Liener (1968) found a relationship between the artificial TL and the exposure age of meteorites that suggested that some of the traps are produced by cosmic-ray irradiation. Thermoluminescence measurements by Vaz (1971b, 1972) and heat flow calculations by Sears (1975b) have shown that only the outer 2-cm surface layer of the meteorites is affected by heating during atmospheric passage. Depth variations in the TL more than 2 cm beneath the fusion crust are not therefore due to this cause. In addition, TL measurements performed by Lalou *et al.* (1970b) on material of the Saint Séverin meteorite placed at different depths inside a target and irradiated with three GeV protons have produced similar depth variations.

The purpose of this study was to analyze the depth dependence of gamma-ray induced TL of several stony meteorites in order to investigate further the changes in the natural TL intensity with depth below the surface of the meteorites. The TL measurements presented here were carried out in samples of Plainview (Texas), Ucera (Venezuela), and Lost City (Oklahoma) meteorites. The Plainview meteorite is a brecciated bronzite found in 1917. Additional fragments were recovered in the nineteen-thirties, and the total amount of material now known is over 700 kg (Hey, 1966). The Lost City meteorite fell January 3, 1970 and four individual meteorites from this

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shower have been found. It was photographed in flight, and recovered by the Smithsonian Prairie Network staff (McCrosky *et al.*, 1971). The five-kg Ucera meteorite fell within two weeks of Lost City on January 16, 1970 (Vaz, 1970). Both, Lost City and Ucera, are homogeneous olivine-bronzite chondrites belonging to type H-5 in the classification of Van Schumus and Wood (Clarke *et al.*, 1971).

EXPERIMENTAL MEASUREMENTS

The samples of Plainview (BM 1959, 805), and Ucera used in this study were obtained from two mutually perpendicular bars removed from 0.4 cm thick slices taken from the completely encrusted stones. The 2 × 4 mm bars of Plainview and the 10 × 4 mm bars of Ucera were then cut into 79, and 56 samples, respectively. The 13 samples of Lost City were removed from a 7.5 cm bar of slice No. 2 of specimen NMNH-4848 of the Smithsonian Institution (Clarke *et al.*, 1971). Figure 1 shows the location of the bars along which the measurements were taken in the three meteorites.

The samples were powdered and sieved, and aliquots of the non-magnetic 50-micron fraction of Plainview, and of the 74 to 177-micron fraction of Ucera and Lost City were used for the TL measurements. The data on Plainview was obtained with a TL analyzer that has been described elsewhere (Sears and Mills, 1974b). The data on Ucera and Lost City was obtained with a Harshaw TL analyzer, model 2000. Duplicate TL measurements run on fresh samples of Ucera with both analyzers showed a maximum deviation between the normalized results of about 7 percent and the average is a few percent. The samples of the three meteorites were heated linearly from room temperature to 500° C at a heating rate of about 5° C per second. The reproducibility of the measurements is ± 5 percent.

Gamma-ray irradiation of the Plainview samples was carried out with a Co-60 source that produced 110 Roentgens per second at the point where the samples were irradiated. A uniform dose of 4.6×10^4 Roentgens was given to these samples after their natural TL was drained by heating to 500° C. The samples of Ucera and Lost City were irradiated with increasing doses of gamma-rays from a Cs-137 source that produced 7.5 Roentgens per second at the center of a 6 cm turntable placed in front of the source. The natural TL of Ucera and Lost City was removed prior to irradiation with Cs-137 gamma rays, with ultraviolet light from low-pressure mercury lamps. Figure 2, curve f, shows the TL curve of Lost City material after u.v. removal of natural TL.

EXPERIMENTAL RESULTS

Very similar natural TL glow-curves are found in the three bronzites that were studied. As shown in Fig. 2, two peaks are present in their natural glow curves; one peak occurs at 230 to 240° C, and the other at 360 to 375° C. The low-temperature peak is more intense than the high-temperature

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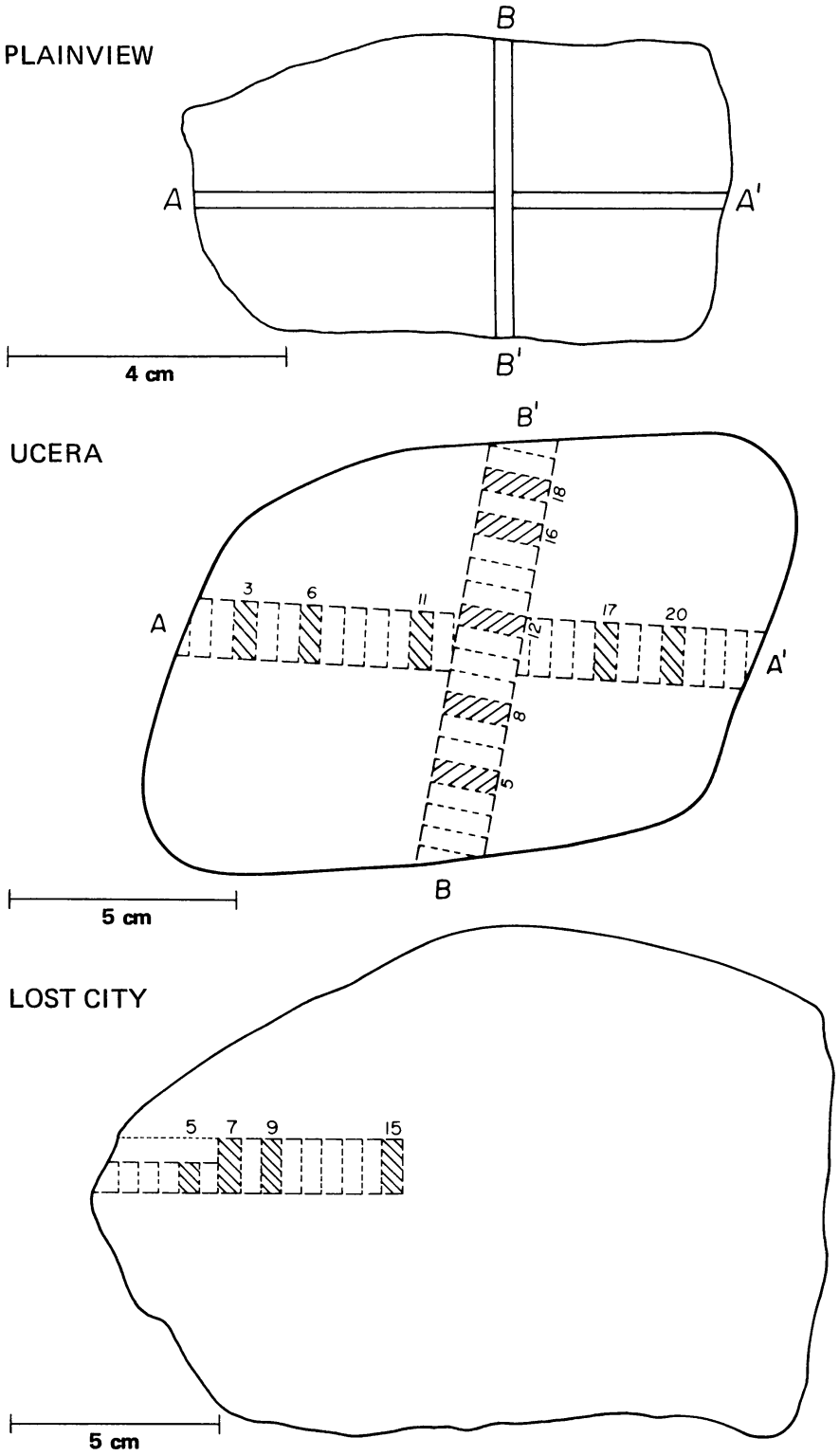

Fig. 1 Diagrams of the slices of Plainview, Ucera, and Lost City showing the position of the bars along which TL was measured. The samples in the bars of Ucera and Lost City shown shaded were the ones used for measurements of artificial TL.

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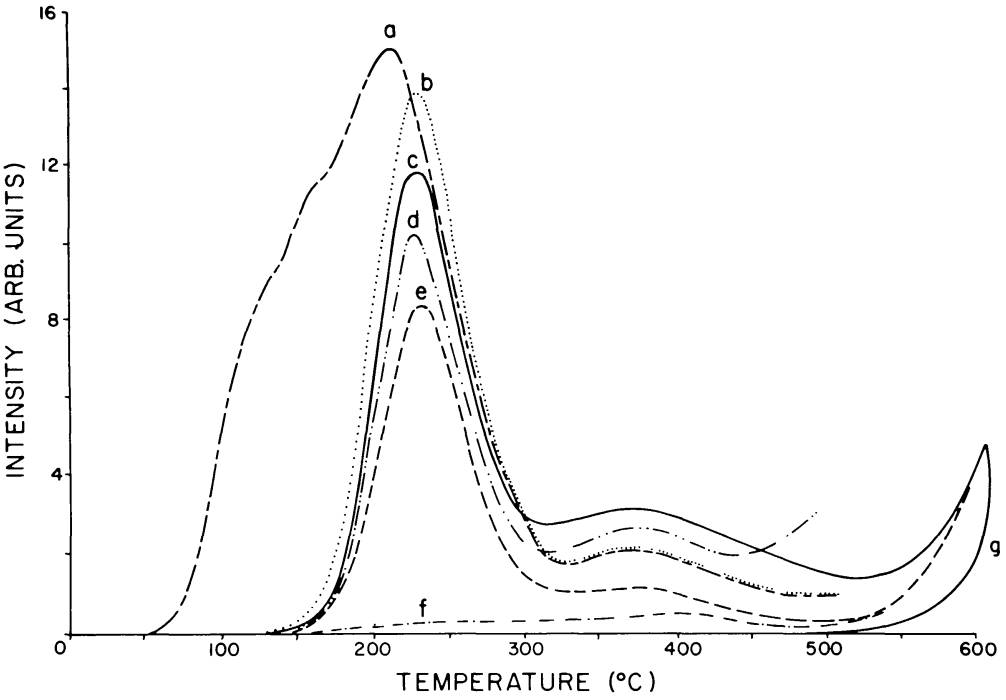

Fig. 2 **TL glow-curves of the meteorites: (a) Lost City after exposure to 5×10^4 Roentgens of Cs-137 gamma rays; (b) same as in (a) but after preheating the irradiated sample at 120° C for two minutes; (c), (d) and (e) natural TL glow-curves of Lost City, Plainview, and Ucera, respectively; (f) Lost City after exposure of a fresh sample to unfiltered ultraviolet light for 48 hours; (g) thermal radiation.**

peak in all three specimens. Irradiation with gamma rays of drained or fresh samples of the stones induces the growth of both natural TL peaks, and of additional glow peaks at temperatures below 200° C.

Figure 3 shows the variation in intensity of the 230° C glow peak of Plainview before and after irradiation with Co-60 gamma rays. The drop in natural TL intensity at the extremities of the two bars is due to heating during the flight of the meteorite. This effect has also been observed in both Ucera and Lost City. At depths greater than about 1 cm from the fusion crust, the natural TL intensity shows a trend in both bars. In the case of the shorter bar, the scatter near the center coincides with a particular xenolith and has been ignored. The artificial TL shows a similar gradient with length. Finally, the ratio of natural to artificial TL shown in the figure has little or no trend, and considerably less scatter than the other two curves because the effect of sample inhomogeneity has been removed. Measurements of the high temperature peak show the same trends but have not been shown for brevity.

Natural TL gradients have been observed previously in the Ucera, and Lost City meteorites (Vaz, 1971a, 1971b). These results are shown in Fig. 4. The plots in the figure show that there is a marked decrease of the natural TL output along bar B-B′ of Ucera and the single bar of Lost City at distances greater than 2 cm from their fusion crusts. In sharp contrast with these results, the natural TL output along bar A-A′ of Ucera is fairly constant except for the already explained drop at the extremities of the bar.

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Figures 5, 6 and 7 show the TL growth curves obtained by irradiating samples of Ucera, and Lost City with Cs-137 gamma rays. These measurements were performed in samples taken from depths greater than 2 cm from the fusion crusts of both specimens: see Fig. 1. To obtain these measurements, different aliquots of each of the 14 samples were illuminated with ultraviolet light for a period of 48 hours to remove the natural TL before irradiating the samples with gamma rays. The irradiation of the u.v.-treated samples with gamma rays induces the growth of the 375° C peak as well as the growth of a broad peak centered at about 150° C in both meteorites. As shown in Fig. 2, the low-temperature peak is made up of at least three over-lapping peaks: one at about 120° C, another at about 150° C, and the glow peak at about 240° C that is present also in the natural glow curves, consistent with earlier work on meteorites (Sears and Mills, 1974b) and lunar samples (Hwang, 1973). Heating the gamma-ray irradiated samples at 120° C for two minutes before TL readout, drains the peaks at 120° C and 150° C leaving only the peaks at 240° C and 375° C populated. This pre-heating period of two minutes at 120° C was used prior to all subsequent TL readouts of artificial TL in Ucera and Lost City. The curves in Figs. 5, 6 and 7 show that the intensity of the gamma-ray induced TL of the samples removed from bar B-B′ of Ucera, and the one bar of Lost City varies from sample to sample according to their depth below the surface of the stones. Furthermore, comparisons of the data in these figures and the TL data in Fig. 4 shows that the relative level of artificial TL of a sample is related to its relative natural-TL level. In other words, the sample with the highest natural TL output in a bar also had the highest artificial TL output. In Fig. 5, the variation in peak intensity among the samples removed from bar B-B′ of Ucera amounts to about 33 percent for the low-temperature peak, and 40 percent for the high-temperature peak. In contrast, the variation in peak intensity in the samples from the bar of Ucera that showed no gradient in natural TL output (bar A-A′) amounts in Fig. 6 to about 17 percent and 14 percent in the low-temperature, and the high-temperature peak, respectively. In the case of Lost City, the plots in Fig. 7 show that the variation in the intensity of the peaks amounts to 26 percent in the low-temperature peak, and 23 percent in the high-temperature peak. The curves in the three figures show also that the high-temperature peak in both meteorites reaches saturation at doses greater than 5×10^5 Roentgens while the low-temperature peak is near saturation only after a dose of about 10^6 Roentgens.

DISCUSSION AND CONCLUSION

According to the very successful model of Randall and Wilkins (1945) for the production of TL, electrons are excited to "traps" in the Conduction Band by any form of ionizing radiation. Thermal energy can dislodge trapped electrons and when they fall to the Valence Band via a luminescent center,

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light is emitted. The intensity of thermoluminescence is, therefore, determined by the number of traps and the number of electrons excited into those traps. The latter is governed by the dose of ionizing radiation received and the thermal history of the specimen, while the former is more complex. In terrestrial samples, traps are usually impurity atoms introduced during crystallization which have been incorporated as interstitial atoms or occupy lattice positions. For example, most Ca-bearing minerals are thermoluminescent because of the substitution in Ca sites. In extraterrestrial materials the possibility also exists that lattice dislocations could be induced as radiation damage and this possibility we examine here.

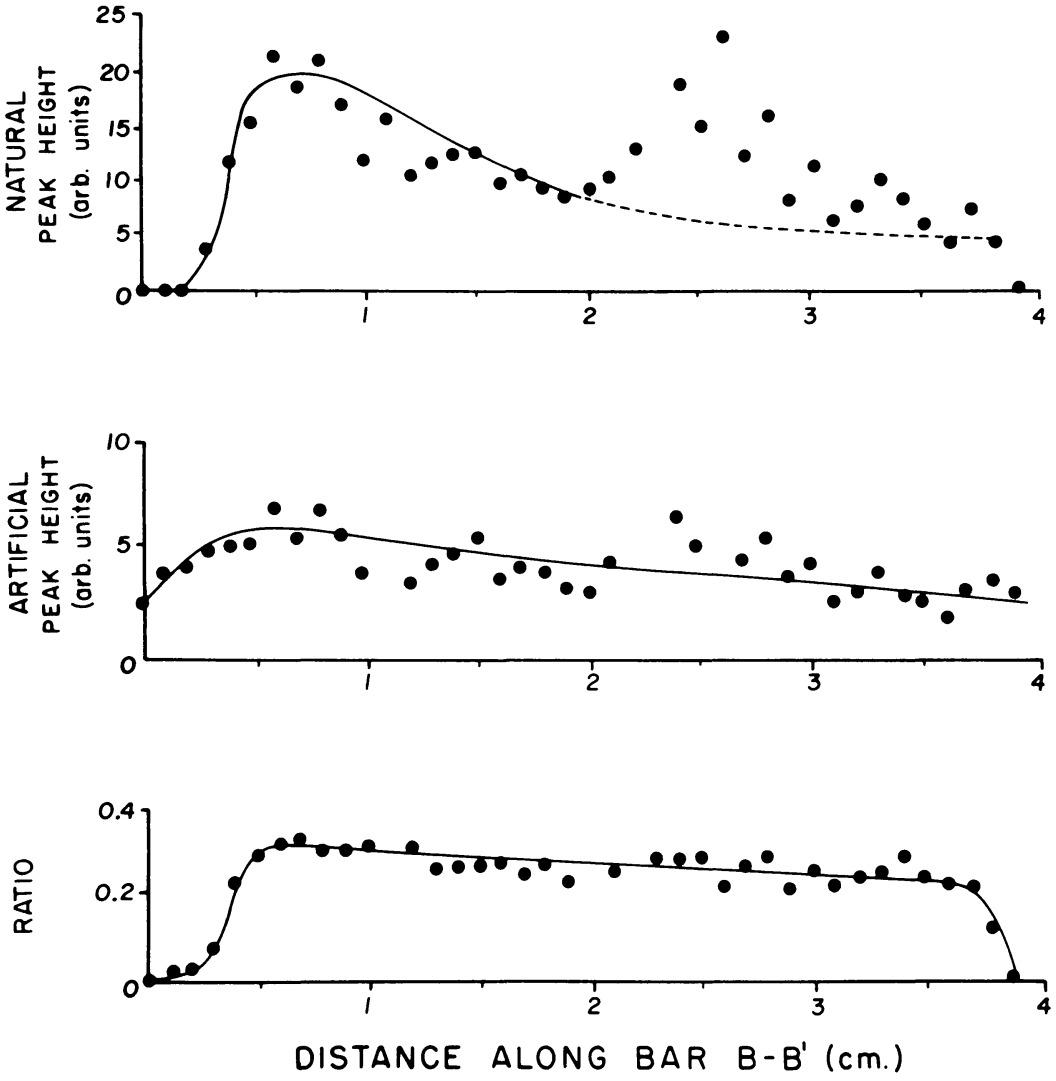

Fig. 3a **Variation in the TL along two mutually perpendicular bars removed from the slice of Plainview. The TL measured is that of the 230° C peak.**

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Natural Thermoluminescence

Measurements of natural TL variations throughout a meteorite reflect the changes in the concentration of trapped charges within the stone. With the exception of the outer 1 cm or so of the meteorite, where atmospheric heating drains the TL (Vaz, 1971a, 1971b; Sears 1975b), the meteorite can be assumed to have had a uniform thermal history, and gradients in natural TL intensity reflect varying degrees of trap population as a result of exposure to ionizing radiation or varying numbers of traps. The sources of radiation available are cosmic rays and long half-life radioactive elements within the meteorite (^{40}K, U, and Th). Experimental and other evidence for the cosmogenic contribution have been presented elsewhere (Sears, 1975a) but may be listed briefly as: simple calculation (Durrani and Christodoulides, 1969); TL exposure age relationships (Houtermans and Liener, 1968); TL – K/Ar age relationships which need to take account of cosmogenic TL (Liener and Geiss, 1968); and finally systematic variations such as those described here and especially, in the large Saint Séverin and Estacado meteorites (Lalou *et al.*, 1970a; Sears, 1975a).

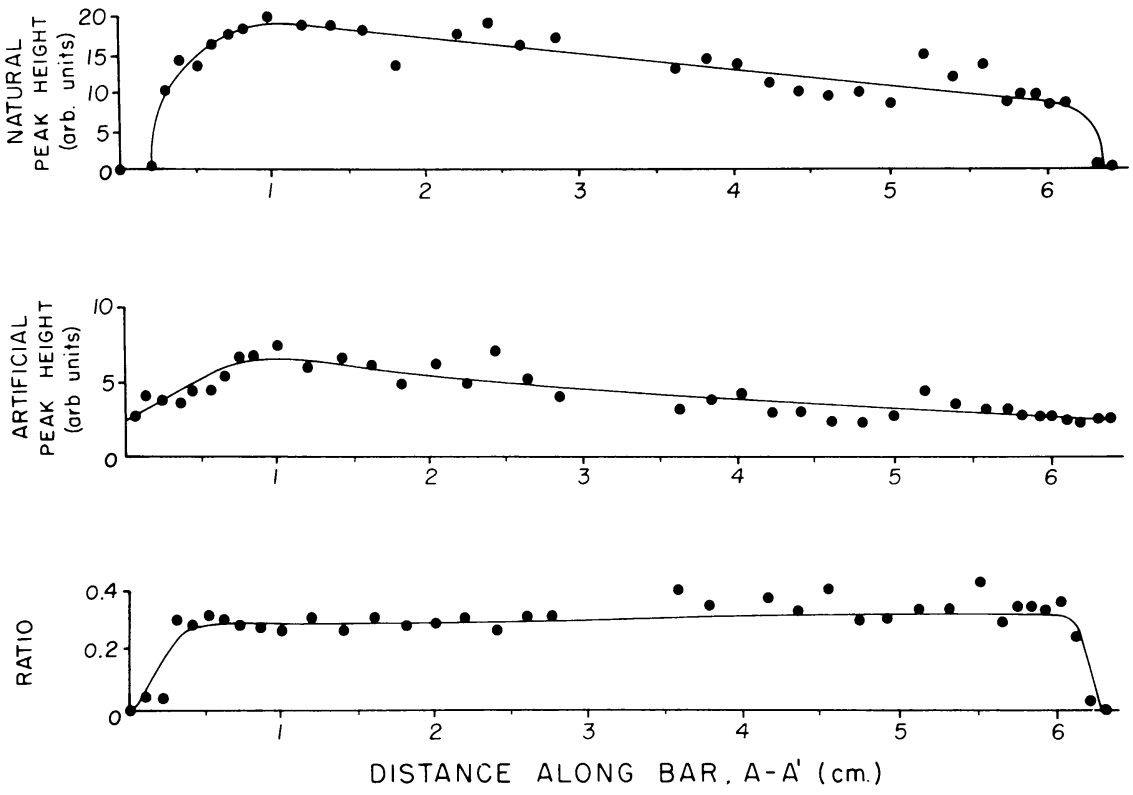

Fig. 3b **See caption on preceding page.**

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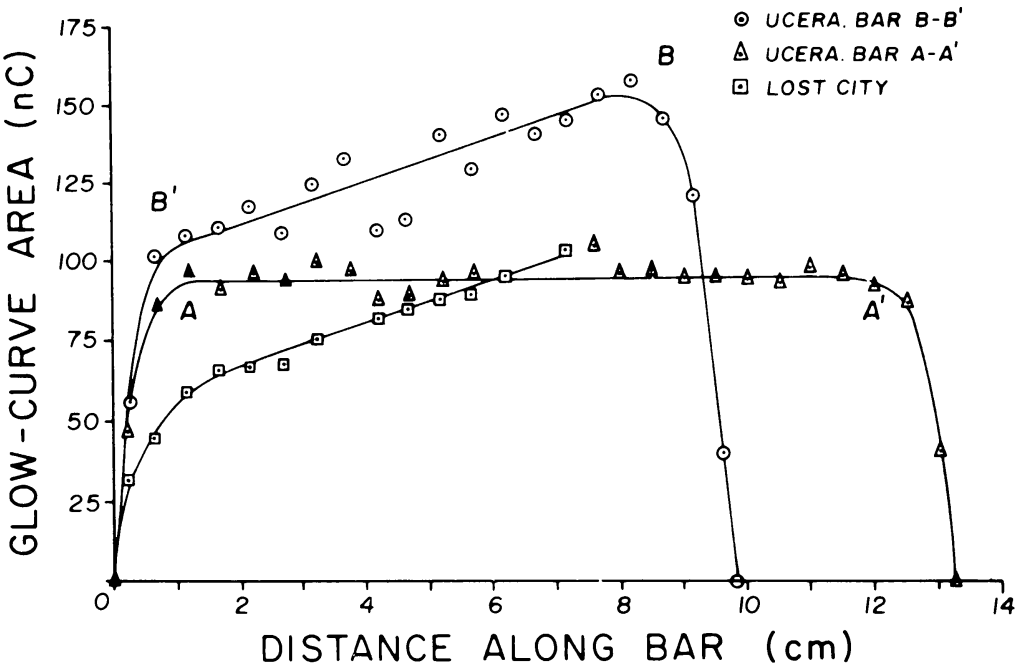

Fig. 4 **Variation in the natural TL output along the two mutually perpendicular bars removed from the slice of Ucera, and the single bar of Lost City that was analyzed.**

It is well established that secondary radiation plays an important part in the production of low-energy spallogenic nuclides. Since TL is a lower energy process (by a factor of 10^3 to 10^6), this is expected to be particularly so (Sears, 1974a) and it has been experimentally observed (Lalou *et al.*, 1970b). As a consequence of cascade of the secondaries, low-energy spallogenic nuclides increase by a factor of ten or so over about 10 cm (for a review see Cameron, 1973), and sometimes more (Wright *et al.*, 1973; Trivedi and Goel, 1973).

One would predict a TL gradient from cosmogenic sources to be similar to that of low-energy spallogenic nuclides. Since radioactive elements are uniformly distributed on the centimeter scale in stony meteorites, one might predict little or no gradient in the TL from this source. However, gamma rays may travel some 10 cm through meteorite material and one cannot exclude the possibility of radiogenic gradients within 10 cm of the preatmospheric surface caused by the internally produced gamma radiation. Fortunately in this connection, it seems unlikely in most cases that material so near the preatmospheric surface would survive atmospheric ablation (Sears, 1975a). Gradients in the natural TL therefore reflect variations in the number of excited electrons and/or the number of electron traps probably resulting from cosmic ray bombardment.

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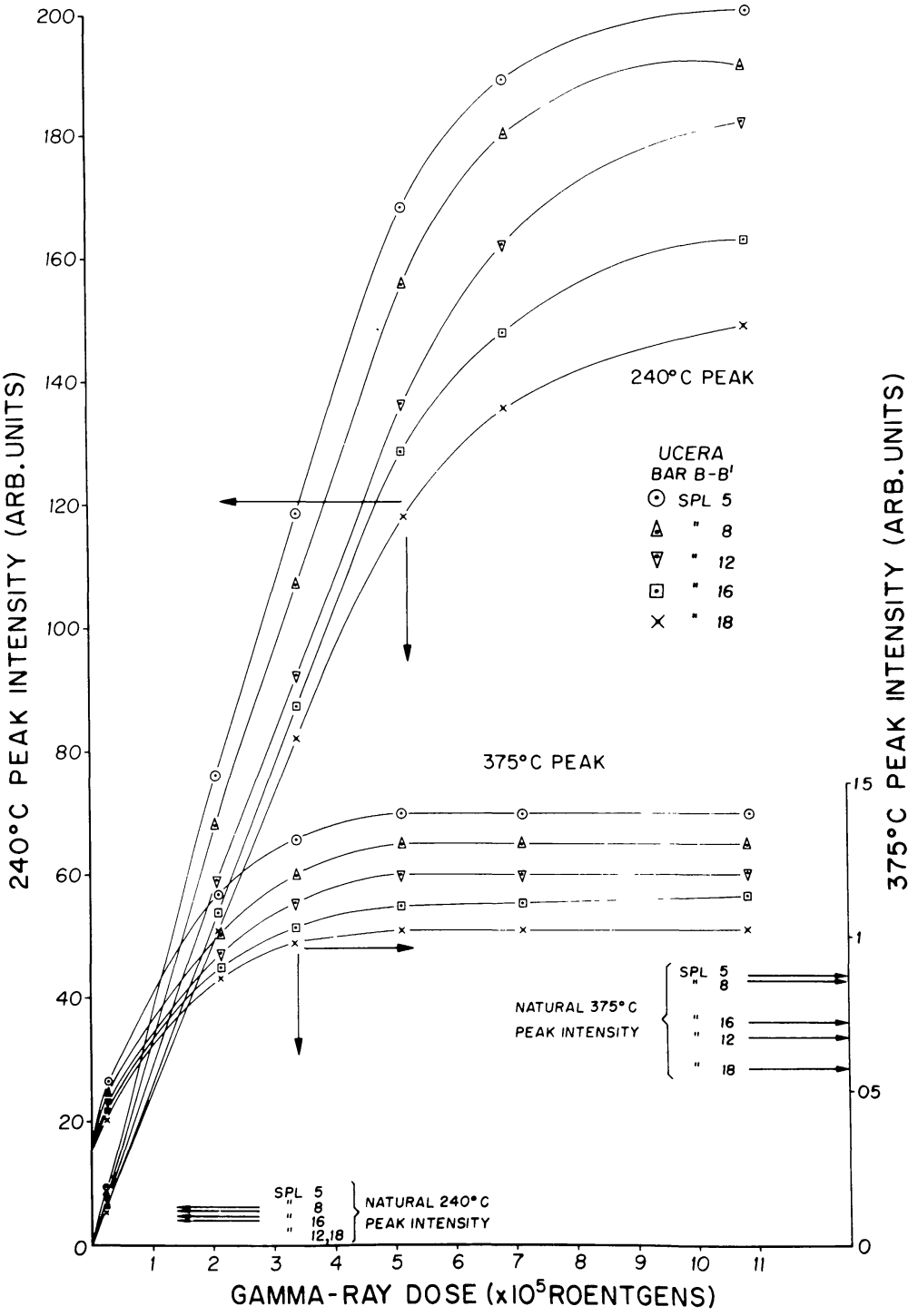

Fig. 5 Peak intensity versus dose curves for the TL induced by exposure to gamma rays in five samples removed from B-B' of Ucera.

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Artificially Created Thermoluminescence

Variations in the numbers of electron traps can be investigated by emptying the traps and subjecting the material to a standard dose of radiation. Two methods are available for the draining process; the material may be heated, or it may be drained with u.v. light. Heating has the advantage of annealing the same material to be used for natural and artificial TL measurements. However, it has poorly understood complications since the process appears to sensitize the material (Christodoulides *et al.*, 1970). The u.v. draining technique works well in removing low-temperature TL but not so the high-temperature TL in the TL-active thickness of the grains. Here we have used both methods with identical results.

Independently we have found that all three meteorites examined show systematic variations in the intensity of the TL created artificially along the previously described bars. Petrographic observations indicate that sample

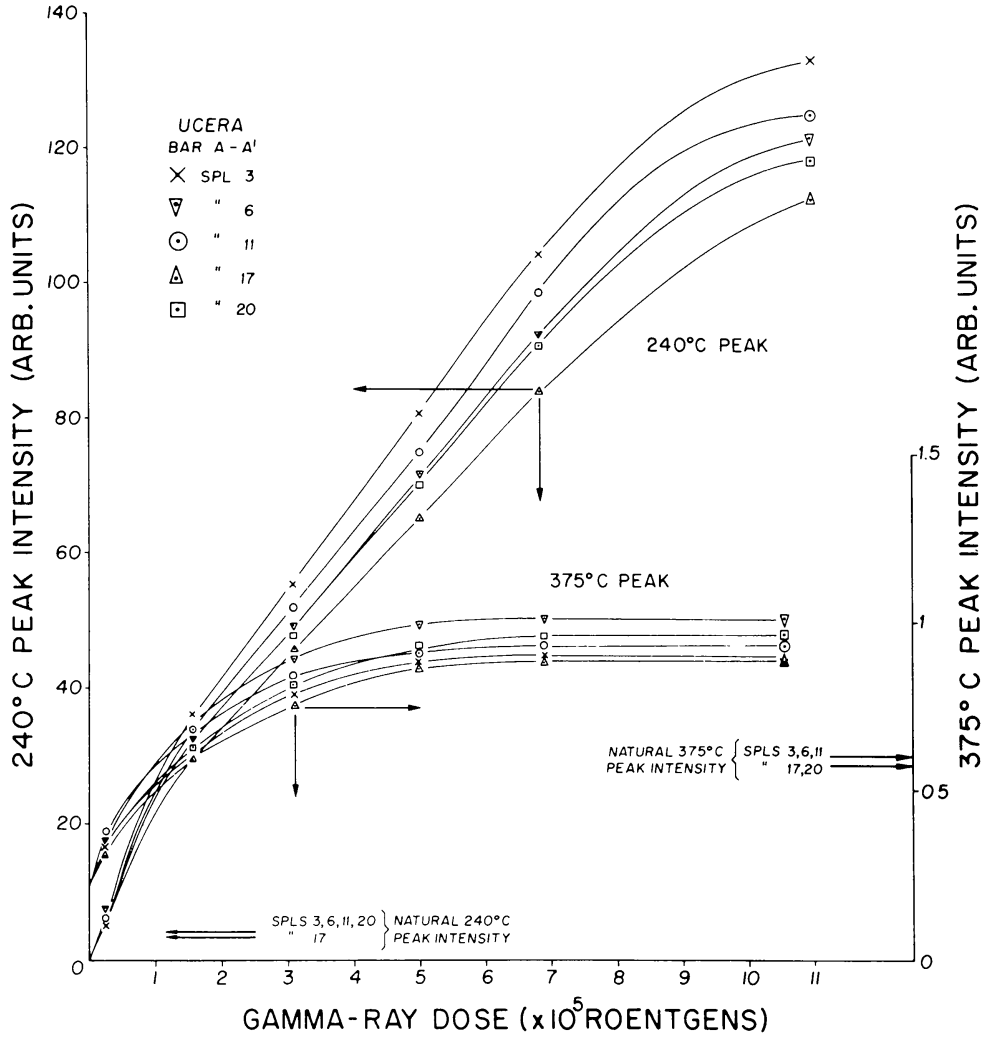

Fig. 6 **Peak intensity versus dose curves for the TL induced by exposure to gamma rays in five samples removed from bar A-A' of Ucera.**

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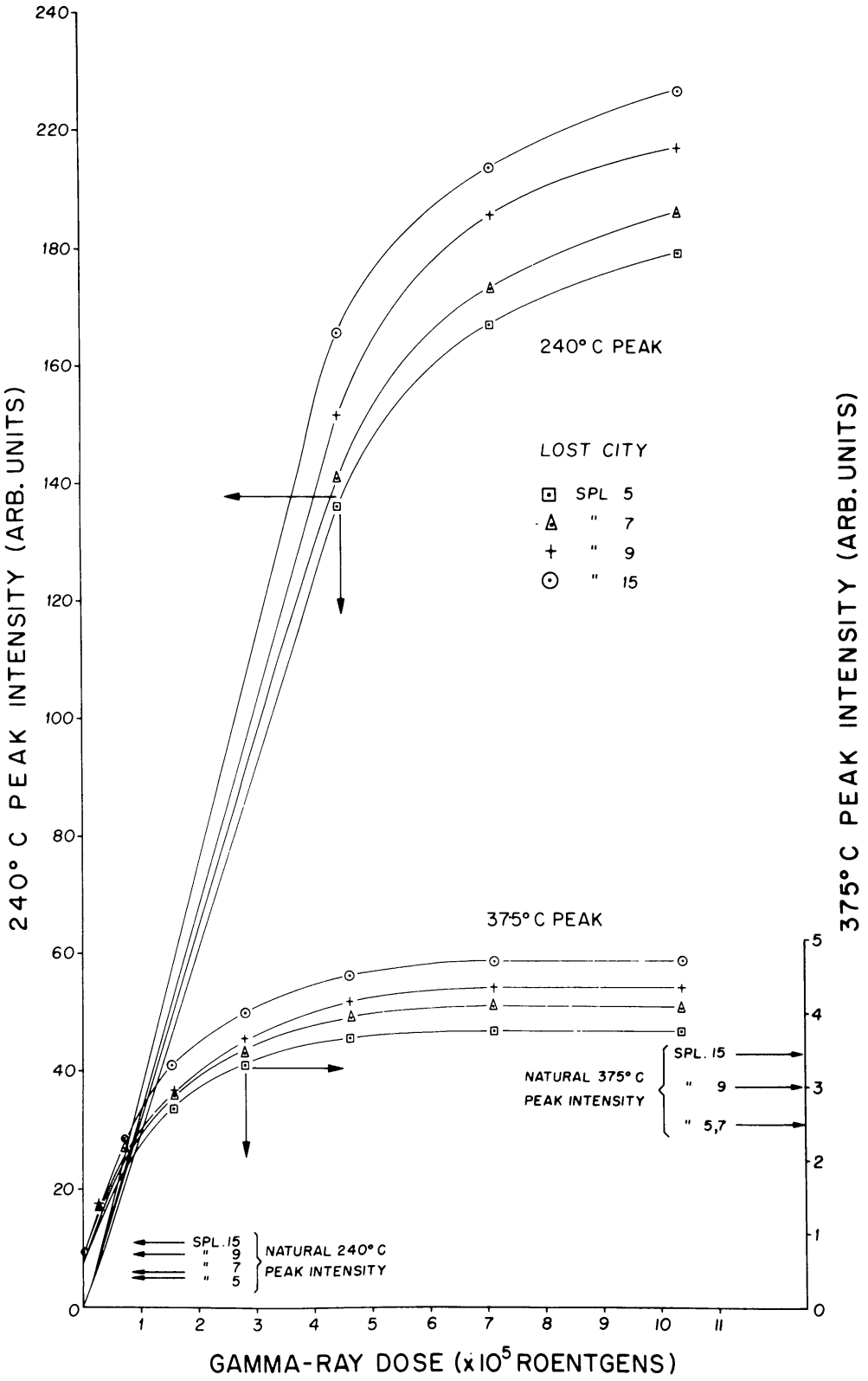

Fig. 7 Peak intensity versus dose curves for the TL induced by exposure to gamma rays in four samples removed from the bar of Lost City.

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inhomogeneity over the order of ten centimeters cannot cause such trends in all three specimens. Similarly detectable temperature gradients do not penetrate the required depths, and for the trends to be caused by internal radioactivity, the specimens would need to come from within the outer 10-cm layer of the meteoroid that is removed by atmospheric ablation. We conclude that the trends are due to systematic variations in trap numbers and are most likely associated with cosmogenic radiation.

It is possible to test experimentally the hypothesis that traps can be produced by radiation damage. Aliquots of five samples of Ucera (three from bar B-B′ and two from bar A-A′) were annealed at 750° C for 24 hours in a nitrogen atmosphere, and irradiated simultaneously with a small dose of Cs-137 gamma-rays (2×10^3 Roentgens). Before TL readout, they were preheated at 120° C for two minutes. The TL induced by the short irradiation of the annealed samples fell within the experimental error of 10 percent for all five samples. By contrast, the difference in TL-producing efficiency between the most and the least efficient sample, when exposed to gamma rays *without* thermal annealing, was a factor of about two. These results seem to indicate that the material recovers from radiation damage with thermal annealing, so that the irradiation with gamma rays can induce only a uniform level of TL in the samples, this uniform level being governed by the number of indigenous *i.e.* nonradiation damage traps. Annealing at 750° C for one hour has been found to be sufficient to remove all fission tracks, which require higher energy particles, from olivine and plagioclase (Fleischer *et al.*, 1970). Plagioclase is the major thermoluminescent mineral in stony meteorites (Lalou *et al.*, 1970a).

We may quote three consequences of the finding of an association between trapping centers and cosmic-ray bombardment for the application of TL to the study of meteorites. Firstly, it shows the need to consider shielding corrections for age determinations by TL, because both the trap populating process and the trap production mechanism may be depth dependent. Secondly, that some caution may be necessary before "normalizing" for sample inhomogeneity by expressing TL as the ratio of natural to artificial; this reservation has been expressed previously by workers on lunar samples (Hoyt *et al.*, 1972). Finally, it suggests a possibility of measuring the TL properties of some large stony meteorites that are too old to retain most of their natural TL, but which may keep in their trap concentration a record of their exposure to cosmic rays.

ACKNOWLEDGMENTS

We are grateful to R. Hutchison of the British Museum, Natural History, for numerous discussions and for supplying the Plainview specimen and R.S. Clarke, Jr., of the Smithsonian Institution, for supplying the Lost City material used in this study.

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Manuscript received 7/29/76

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