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Giant Radiation-Induced Color Halos in Quartz: Solution to a Riddle

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The radii of radiation-induced color halos (RICHs) surrounding radioactive mineral inclusions in mica generally correspond closely to the calculated range of common uranogenic and thorogenic alpha particles in mica. Many exceptions are known, however, and these variants have led investigators to some rather exotic interpretations. Three RICHs found in quartz are identified as aluminum hole-trapping centers. Whereas the inner radii of these RICHs closely match the predicted range of the most energetic common alphas (39 micrometers), the color centers observed extend to 100 micrometers. Migration of valence-band holes down a radiation-induced charge potential might account for enigmatic RICHs. Such RICHs provide natural experiments in ultraslow charge diffusion.

IN 1907 JOLY (*1*) POINTED OUT THAT microscopic color halos commonly observed surrounding small inclusions of radioactive minerals were caused by damage produced by alpha particles emanating from the inclusions. Shortly afterwards, Rutherford (*2*) noted a close correspondence between the radial size of halos and the energies of the alpha particles. A number of workers have described and measured these radiation-induced color halos (RICHs) and, from their sizes, have tried to match them with specific radionuclides in the inclusions [see also (*3*)]. Although it seems possible to relate the sizes of most of the described halos to alpha emitters in the U and Th decay chains, there are many exceptions. Particularly controversial have been two (perhaps artificial) classes of RICHs referred to as Po halos (*3–7*) and giant halos (*3, 8, 9*).

The Po halos are RICHs that have a size and ring structure apparently comparable with the range in silicate minerals of alpha particles emitted by uranogenic Po radioisotopes of mass 210, 214, and 218 (*4–7*), although this interpretation has been challenged (*10*). Significantly, rings that can be attributed to the other five alpha decays in the ^{238}U series seem to be lacking (*4–7*). That the half-life of ^{218}Po is 3 min has not deterred some investigators from proposing separation of Po from its radioactive progenitors before its inclusion in minerals (*4, 5*). Indeed, Po halos have even been offered as possible evidence of an instantaneous creation (*11*).

Giant halos are anomalous RICHs that have radii extending more than approximately 47 μm from edge of the inclusion. Suggested possible mechanisms of formation for these RICHs have included (i) high-energy alphas emitted by rare and undocumented isomers (*9*); (ii) postulated and unknown superheavy elements (*9, 12*); (iii) diffusion of radiolytic atomic H (*13*) and radioactive nuclides through what are structurally and chemically highly anisotropic mineral lattices; (iv) fissionogenic alphas from extinct ^{244}Pu (*14*); and (v) channeling of alphas through open cleavage cracks (*15*).

We have found three giant RICHs surrounding monazite inclusions in quartz grains extracted from two different granites (*16*). These RICHs are seen in reflected light as zones of smoky quartz beginning at a nearly spherical surface 39 μm from the outer edge of the inclusions. Smoky quartz is a common variety of quartz whose gray color is induced by ionizing radiation; the color center is $[AlO_4]^0$, and it is formed as a result of a hole in the valence band being

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trapped at an Al atom that has substituted for Si in the quartz lattice (*17*). The outer radii of the smoky RICHs are 61 to 65 μm for the smallest to 100 to 105 μm for the largest.

These smoky RICHs are in single quartz grains and thus can be viewed from various directions (Fig. 1A); they are close to being spherical. The quartz within a radius of 39 μm of the inclusions is much less strongly colored than are the halos themselves (Fig. 1B). The inner radii of the smoky RICHs are close to the calculated range in quartz of the most energetic of the common alpha particles, the U and Th decay chains (*18*). Radiation effects of alphas in the inner, uncolored volume seem to be of a different type, and this region should exhibit cathodoluminescence (*18*). The outer radii of the smoky RICHs extend far beyond the range of alphas emitted by any known radionuclide but are more than an order of magnitude less than the characteristic range of betas in quartz (*19*). Beginning with the assumptions that the smoky RICHs in quartz are caused by alpha particles radiating from the mineral inclusions and that these alphas are not due to some unknown nuclides or isomers, we have tried to develop an understanding of how such giant RICHs might form.

Aluminum centers are common extrinsic defects in natural quartz. Trivalent Al can substitute for Si, which is tetrahedrally coordinated with O, up to at least several parts per thousand (*20*) without significant overall structural consequences. For each Al, there is then an electron in excess of what is needed to contribute to the local field stability as well as the overall charge balance of the crystal. In natural quartz these substitutional Al atoms are typically compensated by an adjacent cation such as H^+, Na^+, or Li^+ (*21, 22*). Such defects produce no color centers in quartz. The color center in smoky quartz can be produced artificially by ionizing radiation (*17*). Such irradiations elevate electrons to energies of the conduction band, and holes are created in the valence band. Normal room-temperature lifetimes of such electron-hole pairs can be exceedingly short, as they tend to recombine. However, holes and electrons can be trapped at a variety of extrinsic and intrinsic defects in the band gap in quartz where recombination is less likely. A substitutional Al acts as a hole trap with the release of the +1 interstitial cation at temperatures above 200 K (the thermal threshold to drive the cation diffusion along the large *c*-axis channels of the quartz structure) (*21, 23*).

When an alpha decay in the inclusion occurs, the alpha particle starts out with a velocity that depends on its energy. Along its path through quartz, this positively charged mass interacts with orbital electrons. Each interaction transfers some of the energy of the alpha to the electrons. Along most of its path, ionization occurs because of this interaction, and for every valence electron that becomes conductive, a hole is formed. When the alpha has been slowed to the point that it spends enough time in the vicinity of an electron to capture it, a hole is left in the valence band without a balancing conductive electron. The specific ionization strength of an alpha is greatest here, near the limit of its range; a second electron is quickly captured, leaving a neutral He atom and two unbalanced holes.

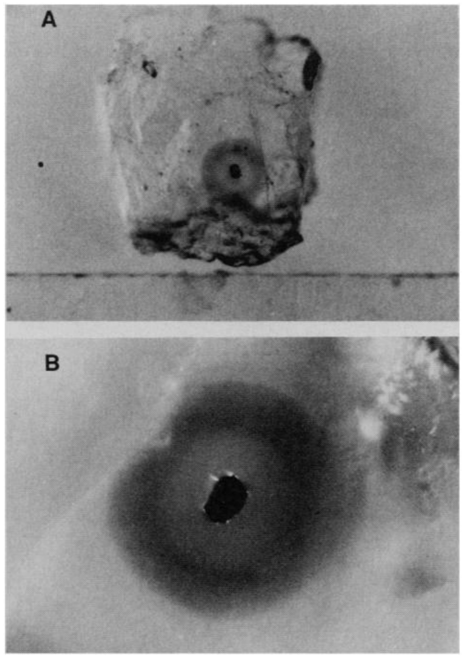

Fig. 1. (**A**) Fragment of a quartz crystal (1.1 mm across) separated from Addaba granite, Lybia, and containing monazite inclusion surrounded by a giant RICH; (**B**) higher magnification of Addaba RICH viewed in reflected light. Outer diameter of smoky halo is 200 to 210 μm.

Thus a narrow zone at the outer limits of the range of alpha particles in quartz is a zone of excess hole production. The continuous production of holes during geologic time drives the electronic equilibrium of this microregime in the direction of stabilizing the Al color centers and producing a thin smoky halo. As hole-capturing centers such as Al defects become saturated, excess holes must migrate outward down a charge potential.

In addition to the 14 alpha particles involved in the decay of ^{238}U and ^{232}Th to Pb, there are typically 10 betas with characteristic ranges in quartz up to 4 mm. Along their path, the betas produce holes and nonvalence electrons in equal numbers. At the limit of their range, betas implant in the quartz lattice free electrons that can combine with holes, be trapped in the band gap at defects, or remain conductive.

Located between a smoky RICH and the characteristic range of betas will be a radiation-induced charge gradient in a natural, semiconducting crystal (*24*). As the excess holes migrate outward under this electric field, some are trapped at more and more remote Al defects. The smoky RICH expands outward, reaching, in one of the two cases we have observed, 105 μm, or more than twice the range of the most energetic alpha particle. It would seem that if smoky RICHs are indeed formed by this mechanism they could potentially grow as large as the range of low-energy beta electrons (*25*).

The relatively low concentration of Al color centers within 39 μm of the inclusion edge (in that volume of quartz that receives nearly the total alpha dose) might be related to the outward migration of holes. However, other factors are also likely important. Studies by electron paramagnetic resonance spectrometry have shown that the concentration of Al hole centers in quartz increases with alpha radiation up to a critical dose level (approximately 1.5×10^7 Gy [1 Gy = 100 rads], although this critical dose will depend in part on the Al concentration), at which point higher doses begin to destroy the Al centers (*26*). Whether this destruction is a result of the conversion of paramagnetic centers to diamagnetic ones, excessive structural damage to the lattice (*27*) or some other mechanism is unknown.

We have no direct measurement of the radioactivity of the monazite inclusion shown in Fig. 1. However, an estimate of the alpha dose experienced by the volume of quartz within 39 μm of a monazite inclusion of known age can be made from typical U and Th concentrations. Such as estimate for the specimen in Fig. 1 yields a total dose exceeding 10^9 Gy (*28*), a dose more than necessary to destroy the Al color centers.

The intensity and size of smoky RICHs in quartz are not simple functions of time and radioactivity of the inclusion but depend on other factors that include (i) the concentration of Al tetrahedra in the quartz lattice, (ii) the annealing characteristics of the color center, (iii) the thermal history of the mineral, and (iv) the physical nature and concentration of other potential hole and electron traps that can affect the electronic equilibria and conductivity. The size of the quartz crystal and conditions external to the crystal surface should have some role in the mobility of conductive electrons and thus some effect on the electronic equilibrium.

Quartz is a mineral whose crystal physics is far better understood than that of mica minerals. Most importantly, the nature of the color center and its response to ionizing radiation is at least partially understood. If anomalous RICHs in other silicate minerals such as micas do not develop in fundamentally different ways (albeit the nature of the color centers can involve defects other than Al), then many of the special conditions and special alpha energies invoked to account for Po and giant halos in mica seem no longer necessary. Giant RICHs can grow by hole diffusion. The apparent absence of ring structures readily associated with U in so-called Po halos might be the result of the destruction of color centers by excessive alpha dose near the inclusions.

Mica is chemically and structurally far more complex than quartz, and there is little understanding of its radiation-induced color centers and carrier behavior. We strongly suspect, however, that the sizes and structure of giant and Po RICHs in mica also are artifacts of radiation-induced conductivity and that their explanation requires neither unknown radioactivity nor an abandonment of current concepts of geologic time.

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28. Average to conservative values for Th and U concentrations in monazites in granites can be selected as 5 and 0.5 weight percent, respectively. For a monazite crystal (density = 5.1 g/cm^3) the size of that in Fig. 1A (approximately 33 by 33 by 46 μm^3), as long as radioactive equilibrium is maintained, alpha decays will deposit annually 0.107 ergs of energy into that mass of quartz within 39 μm of the inclusion (2.15×10^{-6} g), for an annual dose of 497 Gy. The Addaba granite has been dated at 530 million years ago (*29*). The total calculated dose is 2.6×10^{11} Gy.
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Hormonal and Genetic Control of Behavioral Integration in Honey Bee Colonies

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The ability of insect colonies to adjust the division of labor among workers in response to changing environmental and colony conditions, coupled with research showing genetic effects on the division of labor in honey bee colonies, led to an investigation of the role of genetics and the environment in the integration of worker behavior. Measurements of juvenile hormone (JH) titers and allozyme analyses of worker honey bees suggest that two processes are involved in colony-level regulation of division of labor: (i) plasticity in age-dependent behavior is a consequence of modulation of JH titers by extrinsic factors, and (ii) stimuli that can affect JH titers and age-dependent behavior do elicit variable responses among genetically distinct subpopulations of workers within a colony. These results provide a new perspective on the developmental plasticity of insect colonies and support the emerging view that colony genetic structure affects behavioral organization.

ADVANCED INSECT COLONIES HAVE long been likened to "superorganisms" (*1*), a metaphor most apt for traits of colonies that are a consequence of cooperation among individual colony members. One such trait is colony development, which is a consequence of the integration of worker behavior. Results of experimental perturbations (*2, 3*) suggest that insect colonies cope with constant variation in age demography (*4, 5*) and resource availability (*5, 6*) via a process of developmental plasticity that involves ongoing adjustments in the proportions of individual workers engaged in various tasks. The coordination of worker responses to changing environmental conditions is poorly understood. Moreover, the recent discovery (*7–9*) of genetic influences on the division of labor among workers raises new questions about the role of genetics and the environment in integrating activity in insect societies. Regulation of developmental plasticity is a central issue in the study of all biological systems, and comparisons of processes in insect colonies and individual multicellular organisms may provide insights of general significance (*10*).

We describe four experiments that probe the hormonal and genetic basis of developmental plasticity in honey bee colonies. The first two experiments demonstrate that changes in colony age structure can affect age-dependent titers of juvenile hormone (JH) that are associated with changes in worker age-dependent behavior (age polyethism). In the third experiment, treatment with a JH analog affected age polyethism, further supporting the hypothesis that extrinsic factors influence the behavior of worker bees via their effects on JH. The fourth experiment demonstrates genotypic differences in the behavioral responses of workers to altered colony age demography.

JH, a major insect developmental hormone (*11*), is involved in the control of age polyethism in adult worker honey bees (*12–16*). Hemolymph levels of JH increase with worker age (*13, 17*). Low titers are associated with behavior in the nest such as brood

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