calculations[16] show that Coster-Kronig transitions are energetically forbidden when more than two vacancies occur in the outer shell. In the absence of competing Coster-Kronig transitions the fluorescence yield of the L_1 shell increases by more than an order of magnitude. For the present case we estimated that the number of highly ionized atoms ($i > 2$) is too small for a significant effect from the L_1 shell, but more detailed work is needed to study the charge-state dependence of the fluorescence yield and the Coster-Kronig transition rate.

We are very much indebted to Dr. H. Tawara, Dr. K. G. Harrison, Mr. A. Langenberg, Mr. D. Schneider, and Mr. U. Leithäuser who shared in the experimental work. We would like to thank Dr. D. Burch and colleagues for providing us with their experimental results prior to publication, and Professor Dr. W. Mehlhorn for very helpful sugestions. Part of this work is sponsored by FOM with financial support from ZWO.

---

[1] X-ray data: K. G. Harrison, H. Tawara, and F. J. de Heer, to be published; A. Langenberg and J. van Eck, to be published [using the same technique as F. W. Saris and D. Onderdelinden, Physica (Utrecht) 49, 441 (1970)]. Auger data: N. Stolterfoht, D. Schneider, and P. Ziem, to be published.

[2] Saris and Onderdelinden, Ref. 1.

[3] D. J. Volz and M. E. Rudd, Phys. Rev. A 2, 1395 (1970).

[4] R. K. Cacak, Q. C. Kessel, and M. E. Rudd, Phys. Rev. A 2, 1327 (1970).

[5] D. Burch, W. B. Ingalls, J. S. Risley, and R. Heffner, private communication.

[6] F. P. Larkins, J. Phys. B: Proc. Phys. Soc., London 4, L29 (1971).

[7] C. P. Bhalla and K. L. Walters, in *Proceedings of the International Conference on Inner Shell Ionization Phenomena, Atlanta, Georgia, 1972,* edited by R. W. Fink, S. T. Manson, I. M. Palms, and P. V. Rao, CONF-720 404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973), p. 1572.

[8] *Proceedings of the International Conference on Inner Shell Ionization Phenomena, Atlanta, Georgia, 1972,* edited by R. W. Fink, J. T. Manson, I. M. Palms, and P. V. Rao, CONF-720 404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973).

[9] F. W. Saris, in *Proceedings of the Seventh International Conference on the Physics of Electronic and Atomic Collisions. Invited Talks and Progress Reports,* edited by T. R. Govers and F. J. de Heer (North-Holland, Amsterdam, 1972).

[10] N. Stolterfoht, Z. Phys. 248, 81,92 (1971).

[11] K. G. Harrison, H. Tawara, and F. J. de Heer, to be published.

[12] A. Langenberg and J. van Eck, to be published.

[13] W. Mehlhorn, Z. Phys. 208, 11 (1968).

[14] W. Mehlhorn and D. Stahlherm, Z. Phys. 217, 294 (1968).

[15] F. P. Larkins, J. Phys. B: Proc. Phys. Soc., London 4, 1 (1971).

[16] W. Mehlhorn, private communication.

Energetic Heavy-Particle Detection by Photochromic Material

P. J. Ouseph
Physics Department, University of Louisville, Louisville, Kentucky 40208

(Received 20 February 1973)

Mechanisms are discussed by which radiations can produce color changes in Fe-doped $SrTiO_3$. α-particle and cosmic-ray (π-meson) absorption events have been detected by these crystals.

We report here the detection of high-energy particles by photochromic iron-doped strontium-titanate single crystals. This method is much simpler than detection by other presently existing track-type detectors. As explained below, the tracks can be erased and the crystal may be reused.

Strontium-titanate crystals doped with 0.1% iron are black in color. The iron ions enter the crystal substitutionally, replacing Ti^{4+}. About half of the iron ions (~ 52%) are in the Fe^{4+} state.[1] The black color is explained in terms of the high absorption coefficient of Fe^{4+} in the visible region of the spectrum.[2] If such a crystal is reduced by heating at 900°C for about $\frac{1}{2}$ hour in partial vacuum (1 Torr), the number of Fe^{3+} ions increases and the crystal changes color from black to yellow. Since the Fe^{3+} ions are in the Ti^{4+} sites, charge balance in the crystal is maintained by oxygen vacancies.[3] By changing the valence state of the iron ions, the color of the crystal can be switched back and forth between black and

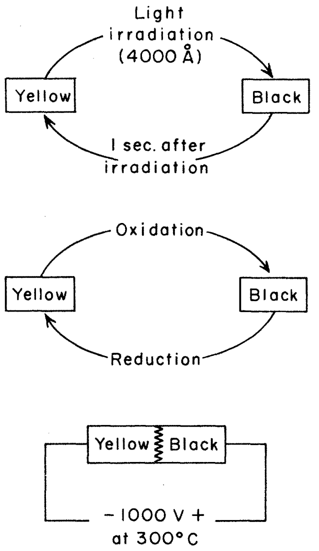

FIG. 1. Processes that produce reversible color changes in $SrTiO_3$.

yellow.

In Fig. 1 are shown three methods by which the color can be changed from yellow back to black. They are (1) light irradiation in the range 3900–4300 Å, (2) oxidation, and (3) application of high voltage across the crystal above 100°C. In the first method, an electron is raised to the conduction band by the incident photon producing the valence change from Fe^{3+} to Fe^{4+} without any change in the oxygen vacancies. Subsequently, the electron falls back to the iron ion site with a decay time of less than 1 sec; therefore, the color change produced by light irradiation is not stable. The other methods depend on increasing or decreasing the oxygen vacancies and, therefore, they produce stable states which can be restored to the original form (i.e., black to yellow) by reversing the process. The method that is significant to the discussion here is the third one where, under the influence of the applied electric field, O^{2-} ions move to the positive electrode side when heated above 100°C, thereby increasing the Fe^{4+} concentration and, consequently, the optical density near this electrode.[4] Thus a visible color change (blackening) occurs near the positive electrode since the mobilized oxygen ions result in filling the oxygen vacancies.

When certain radiation particles pass through a crystal, the core of the track extending out to a given radius causes momentary local heating along the track. If the crystal during this time is subjected to an electric field, the oxygen ions are attracted to one side of the particle track, producing a black image in regions where temperatures have exceeded about 100°C. Observability of this black region depends on the radius of the effective particle track. A simple calculation using the method of Mullen[5] shows that, for particles having energy losses in $SrTiO_3$ no greater than 15 MeV/mg cm^2, the effective diameter of the region heated to 100°C is <1000 Å.[6] This track size is too small for optical microscopic detection. Results of experiments performed to detect 40-MeV α particles using a 25-mm-diam $\times$2-mm strontium-titanate crystal support the above conclusion. The crystal was subjected to an applied electric potential of 1000 V, and the particles entered the crystal through its edge. Even though single tracks of the 40-MeV α particles were not made visible microscopically by this technique, a general blackening of the crystal occurring in a well-defined region (whose width was equal to the range $\sim$0.5 mm of the α particles in $SrTiO_3$) was observed on irradiating the crystal in the cyclotron beam. This blackening was stable. The mechanism for it is different from that observed by Faughnan, Phillips, and Kiss[7] in which the crystal was bombarded by 20–50-keV electrons. An electric field was not applied during their irradiation. The blackening obtained by them resulted from electronic transitions to the conduction band, producing Fe^{4+} (similar to method 1). In this case, the electrons fall back to an original state in a characteristic decay time, thereby restoring rather rapidly the original color of the crystal.

An experimental approach that yielded satisfactory single-particle detection is as follows: Experimentally (see Fig. 2), 2-cm-radius$\times$2 mm-thick strontium-titanate crystal (yellow) was sandwiched between two uranium foil electrodes, one of which (anode) had a thin oxide layer ($\sim$0.1 mm) on it. When α particles emitted during the decay of uranium pass through the oxide layer, they produce oxygen ions which are attracted to the $SrTiO_3$ crystal by the applied electric field (1000 V between the electrodes). An oxygen accumulation on the surface of the crystal becomes visible as a result of the formation of clusters of Fe^{4+} sites. Projection of the cross section of the α-particle tracks thus obtained are shown in Fig. 3. Spots having a diameter of $\sim$10 μm are formed. The range of these α particles in uranium oxide is $\sim$10 μm; therefore, most of these particles are stopped in the oxide layer. The reasons for suggesting that oxygen ions released from the

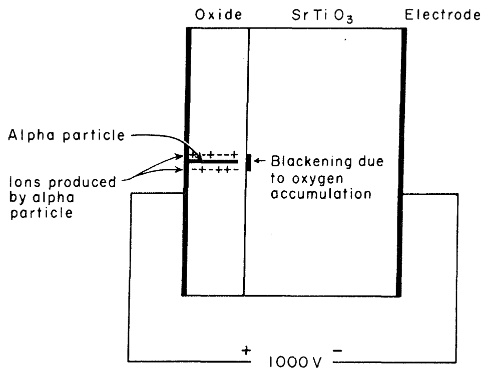

FIG. 2. Experimental arrangement used in detection of radiation. α particles release positive oxygen ions in the uranium-oxide layer. The positive oxygen ions are attracted to $SrTiO_3$ and oxygen accumulation produces color changes on the surface of the crystal. The depth of the blackening is too small to measure. Thickness of the $SrTiO_3$ crystal is 2 mm and the thickness of the oxide layer is ~ 0.1 mm.

oxide are responsible for the spots are (1) spots form on the surface of the crystal facing the anode coated with the uranium-oxide layer. (2) The spot size observed is much larger than the calculated radius of the radiation heated region around the particle-track core. (3) Uranium foils without one having an oxide layer did not produce any images.

Figure 3 also shows a long track ending in a three-pronged star which occurred during one of the experiments. This track starts from the top edge of the crystal, and is presumed to be caused by a cosmic-ray particle being slowed down in the oxide layer. A π^- meson captured by a nucleus after being slowed down produces similar tracks in photographic emulsion.

The tracks can be erased by reducing the crystal at 900°C in partial vacuum. The number of such reducing cycles has no effect on the sensitivity of the crystal. Crystals which have been reduced up to 10 times have been reused successfully. Heating at 900°C also apparently restores the crystal from radiation damages which may have caused such effects as small changes in volume and dielectric constant.[8] Other advantages are these: (1) Image development is immediate requiring no further special methods. (2) Temperature or pressure control are unnecessary. (3) Unlike a cloud or bubble chamber, the sensitivity is continuous. (4) The image is stable, permitting leisurely analysis of results.

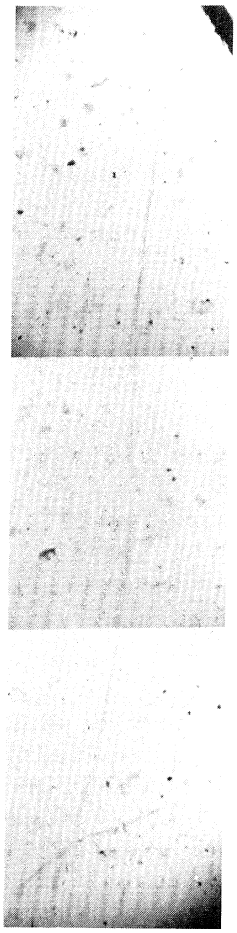

FIG. 3. Spots are projections of α-particle tracks on the surface of the $SrTiO_3$ crystal. Most of the α particles emitted during the decay of uranium are stopped in the oxide layer. The range of these α particles in the oxide is ~ 10 μm. The long track is apparently due to a cosmic-ray particle being slowed down and reacting with a nucleus in the oxide layer. The total length of the track is 4.6 mm.

In the arrangement just described, the active volume of the detector is very small, being confined to a small section of the oxide layer in contact with the crystal.

The author is grateful to B. W. Faughnan of the RCA Laboratories, Princeton, New Jersey, N. Nolan, Michigan State University, and K. L. Gauri, D. Dupré, and H. Groskreutz for many helpful discussions.

---

[1] C. T. Luiskutty and P. J. Ouseph, Bull. Amer. Phys. Soc. 17, 261 (1972). The amount of Fe^{4+} (52%) is estimated from Mössbauer absorption spectra. In reduced

samples there are about 80% Fe^{3+} and 20% Fe in lower valence state. Fe^{4+} has also been observed in photochromic TiO_2: A. J. Nozik, J. Phys. C: Proc. Phys. Soc., London 5, 3147 (1972).

[2] B. W. Faughnan, Phys. Rev. B 4, 3623 (1971). Relationship between color changes and valence states of iron is also discussed in the following articles: B. W. Faughnan and Z. J. Kiss, Phys. Rev. Lett. 21, 1331 (1968); K. A. Müller, Th. von Waldkirch, W. Belinger, and B. W. Faughnan, Solid State Commun. 9, 1097 (1971); E. S. Kirkpatrick, K. A. Müller, and R. S. Rubins, Phys. Rev. 135, A86 (1964). Also see J. Blanc and D. L. Staebler, Phys. Rev. B 4, 3548 (1971).

[3] In Fe-Mo-doped strontium-titanate crystals charge balance is maintained by the higher valence of Mo and, therefore, no oxygen vacancy is required. Isomer-shift measurements have supported this view. The difference in isomer shift of Fe^{3+} in Fe-doped and Fe-Mo-doped crystals can be explained in terms of the change in the overlap contribution to the electronic density at the nucleus due to the change in the number of oxygen ions surrounding Fe^{3+} (five oxygen ions and a vacancy in singly doped crystals and six oxygen ions in doubly doped crystals).

[4] Blanc and Staebler, Ref. 2.

[5] J. G. Mullen, Phys. Lett. 15, 15 (1965).

[6] H. E. Groskreutz and P. J. Ouseph, to be published.

[7] B. W. Faughnan and Z. J. Kiss, IEEE J. Quantum Electron. 5, 17 (1969).

[8] H. Weik, Phys. Lett. 9, 92 (1964); G. K. Miner, thesis, University of Cincinnati, 1965 (unpublished); S. P. Solov'ev, I. I. Kuzmin, V. V. Zakurkin, and V. Ja. Dudarev, J. Phys. (Paris) 33, 443 (1972).

[9] $SrTiO_3$ crystals were obtained from the National Lead Co., South Amboy, New Jersey. These crystals are consistent in their color switching properties. The coloring is a volume effect and is independent of the axes of the crystal.

Phenomenological Theory of Collisionless Superfluid ^{3}He

S. J. Putterman*
Physics Department, University of California, Los Angeles, California 90024

(Received 16 April 1973)

According to a phenomenological theory of collisionless superfluid ^{3}He, the velocity of high-frequency sound should show a transition from zero- to first-sound velocities as the superfluid fraction ρ_s/ρ is increased. From the experimental observations of this behavior by Paulson, Johnson, and Wheatley, the order parameter ρ_s/ρ can be determined for ^{3}He II.

Observations of a discontinuity in the specific heat[1] and a shift of the NMR frequency[2] in pressurized ^{3}He near 2-3 mK have raised the possibility that it undergoes a superfluid transition.

In the normal state, ^{3}He displays interesting collective or macroscopic behavior in two regimes: (1) the hydrodynamic regime, where the angular frequency ω of any disturbance is small compared with $1/\tau$ (τ is the collision time), and (2) the collisionless regime where $\omega\tau > 1$. If ^{3}He exhibits superfluidity, there will thus be two corresponding regimes in which the superfluid fraction will influence the collective behavior. It is generally expected that in the superfluid-^{3}He hydrodynamic regime the appropriate phenomenological theory will be the Landau two-fluid equations[3] of ^{4}He supplemented by the Onsager-Feynman quantization

$$\oint \vec{v}_s \cdot d\vec{l} = nh/2m_3, \tag{1}$$

where $\vec{v}_s$ is the superfluid velocity, n an integer, h Planck's constant, m_3 the mass of a ^{3}He atom, and $d\vec{l}$ an element of length in the contour of integration.

The purpose of this paper is to present a phenomenological theory of the collisionless regime of superfluid ^{3}He. It will be seen that as a result of the formation of a superfluid the speed of high-frequency or zero sound must, as the temperature is lowered, show a transition to the first-sound velocity. This behavior has already been observed by Paulson, Johnson, and Wheatley[4] and a comparison of the results obtained here with their experiments leads to an expression for the superfluid fraction ρ_s/ρ.

The excitations or normal fluid are completely described by the distribution function $n(\vec{r}, \vec{p}, t)\, d^3r \times d^3p$, yielding the number of excitations in the phase-space region d^3r about $\vec{r}$ and d^3p about $\vec{p}$. As with ^{3}He in the normal state,[5] we assume that $n(\vec{r}, \vec{p}, t)$ obeys a kinetic equation of the form

$$\frac{\partial n}{\partial t} + \frac{\partial n}{\partial r_\alpha}\frac{\partial \epsilon}{\partial p_\alpha} - \frac{\partial n}{\partial p_\alpha}\frac{\partial \epsilon}{\partial r_\alpha} = 0, \tag{2}$$

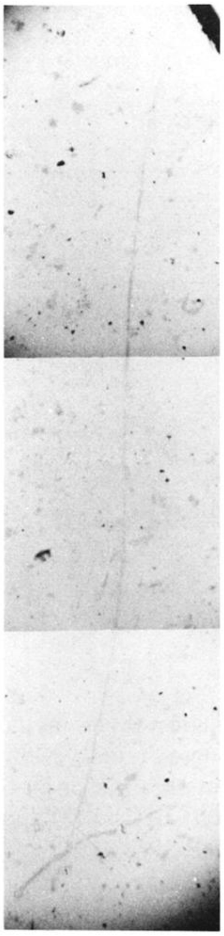

FIG. 3. Spots are projections of α-particle tracks on the surface of the $SrTiO_3$ crystal. Most of the α particles emitted during the decay of uranium are stopped in the oxide layer. The range of these α particles in the oxide is $\sim 10\,\mu m$. The long track is apparently due to a cosmic-ray particle being slowed down and reacting with a nucleus in the oxide layer. The total length of the track is 4.6 mm.