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Hyperconductivity in fluorphlogopite at 300 K and 1.1 T

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Abstract – We report on studies of hyperconductivity in which electric charge moves in a solid in the absence of an applied electric field. This is indicative of a non-Ohmic mechanism. Our results are consistent with charge being carried ballistically by mobile nonlinear lattice excitations called quodons moving along close-packed atomic chains in the cation layers of some silicates. The finding that quodons can trap and carry a charge was first found by the authors in muscovite (RUSSELL F. M. *et al.*, *EPL*, **120** (2017) 46001), which previously was not possible. In this paper we have also found hyperconductivity in lepidolite, phlogopite and synthetic fluorphlogopite but not in biotite or quartz. We have found that a current continues to flow for many seconds after the creation of quodons is stopped, indicating they have long flight-paths. This shows that quodons are decoupled from phonons, must experience elastic reflection at boundaries and are not stopped by inevitable dislocations or other minor defects. We have also found that quodons can anneal defects caused by mechanical working of crystal faces. The current carried by quodons is unaffected by a magnetic field of 1.1 T.

Introduction. – The transport of localized packets of energy along the close-packed chains of muscovite mica was observed experimentally by one of the authors by bombarding one side of a single crystal with alpha particles and detecting the ejection of atoms at the remote opposite side along the lattice directions in the potassium layer [1]. Charge transport in the absence of an electric field was experimentally demonstrated by some of the authors in small natural crystals of muscovite [2] and was called hyperconductivity (HC) to distinguish it from superconductivity. Here we report that hyperconductivity has been demonstrated in several other silicates and not only in muscovite. A necessary but not sufficient condition is the layered structure of phyllosilicates. The proposed explanation is that the electric charge could be coupled to mobile nonlinear lattice excitations that travel ballistically and are called quodons [3]. The new experiments presented here have allowed further study of hyperconductivity and quodon properties.

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The background to these experiments was an extended study of fossil tracks in muscovite crystals created by charged particles emitted during radioactive processes [4–7]. It was found that the nuclear recoil from β^- decay created a nonlinear lattice excitation, called quodon, that bound with, and carried away, the positive charge left behind by the emitted electron [8–10]. Measurements showed that the extent of decoration on fossil tracks associated with quodons carrying charge was the same as that on a slowly moving positron [6–8]. This related with a previous study of charge binding to discrete breathers [11,12].

For the commonest 2M1 polymorph the lattice has a unit cell of about 84 atoms, which makes modelling of nonlinear lattice excitations difficult. As a result, most theoretical and numerical studies of possible excitations in the potassium sheets use simplified quasi-2D models or even 1D chains, showing mobile lattice excitations, such as solitons, breathers and kinks [13–16].

However, the observed creation of multiple secondary mobile excitations seen in the fossil tracks are indicative of breather-like excitations because of their smaller energies. Numerical modelling in 2D have simulated breather propagation along chains for up to 10^6 atoms [17] and ejection of an atom from the surface in a 1D model [18].

The interplay of lattice excitation with electrical charge has a long history leading to polarons, solectrons, polarobreathers, solitons and kinks [19–28]. Electric currents that are independent of an electric field were first observed in polydiacetylene, with carriers created by photo-absorption [29]. These properties are reported to be of interest in microelectronics for providing components with extremely small dissipation [30].

The finding of crystals of muscovite with fossil tracks consistent with quodons that can transport a single positive charge over great distances, up to 0.5 m or 10^9 atoms, in the absence of an applied voltage led to the prediction of hyperconductivity, which was verified by experiment [2,31]. Beta decay of the ^{40}K isotope leads to the build-up of reservoirs of charge. Holes tend to concentrate in or near the potassium sheets with electrons more distributed in the lattice. The high-volume resistivity of muscovite hinders their recombination.

Injection of an energetic alpha particle into muscovite creates many thousands of quodons and phonons but injects at most two holes. When alpha irradiation starts on a previously non-irradiated crystal, there is initially a large current that greatly exceeds the current of holes fed in by the alpha particles. This can only be due to the presence of the quodons as muscovite is an excellent insulator. This initial peak current was predicted as a critical feature of charge transport by quodons and was confirmed by experiment [2]. The initial current decreases with time and tends to a limiting value that is determined, in part, by the influx of holes by the alpha particles. There can also be a component to this limiting current from normal conduction, both surface and bulk, due to applied or induced voltages provided by a non-uniform distribution of charge [32].

In this article, evidence is presented that quodons can be elastically reflected at boundaries, can transport charge across a crystal interface, can anneal crystal damage and are not affected by a magnetic field of up to 1.1 T. These results are presented in different subsections of the section "Results", which is preceded by the description of the equipment in the section "Equipment" and followed by a discussion of the results and their implications in the section "Discussion". The article finishes with the "Conclusions".

Equipment. – The experiment built on the techniques developed in the demonstration of particle ejection by quodons [1] but extended it to allow measurement of currents instead of counting individual particles [2]. Weak sources of alpha particles again were used because of their compactness and availability to create neutral quodons. An alternative method is to use a plasma or ion beam [33]. A radioactive source has the advantages that it does not create electrical noise and can be used for metals or insulators. The apparatus used consists basically of a target crystal that is bombarded with alpha particles at one point and is connected at a remote point to a current amplifier feeding a data logger. The alpha source consisted of a linear array of seven ^{241}Am disc-foils each nominally rated at 1 μCi, giving 3.7×10^4 alphas/s. On average, an alpha creates more than 10^4 quodons with energies of up to about 10 eV but injects only two units of positive charge. The schematic arrangement is shown in fig. 1.

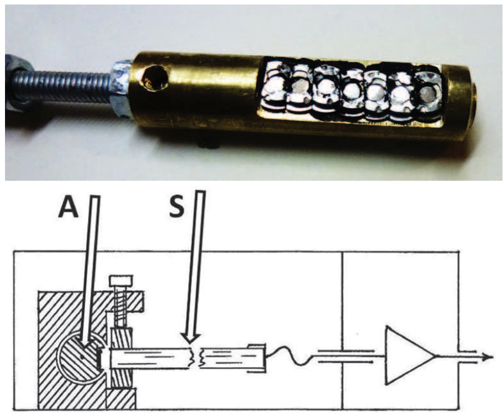

Fig. 1: Top: photo of the ^{241}Am alpha-particle source. Bottom: schematic diagram of the experiment. The photo shows seven alpha-emitting foils bonded to a brass rod. This is inserted into a brass block that has a slot cut parallel to the axis of rotation of the brass rod. The rotation of the rod controls the number of alphas passing through the slot. This arrangement is shown in cross-section at the left of the diagram marked (A). The mica strip (S) is placed near to the slot with the other end held in a metal terminal connected to the current amplifier shown at the right of the diagram.

To minimise interference from secondary electrons and sputtered ions, the target face and metal clamp are covered with gold leaf. This causes about a 20% drop in the flux of alphas particles. This is further reduced by the geometry of the source and holder to give a maximum available current of around 40 fA. To minimise contact potentials, gold foil also was used for electrical contacts to the target at both the clamp and remote terminal. The strip or slab of the target crystal and the surrounding box formed a capacitance of about 2×10^{-10} F.

The current was measured with a FEMTO DDPCA-300 sub-femtoampere current amplifier[1]. In isolation under open-circuit conditions this unit indicated a rms noise of approximately 0.5 fA. The rise time of the amplifier was set at 0.5 s. The output from the amplifier was fed to a MadgeTech Volt101A data logger for storage and visual display to enable real-time study of variables[2]. To minimise line-borne noise, the amplifier was fed by an uninterruptable power supply.

[1]Manufactured by FEMTO Messtechnik GmbH, `http://www.femto.de/en`, accessed February 2019.

[2]Manufactured by MadgeTech Inc. Warner, USA, `https://www.madgetech.com/`, accessed February 2019.

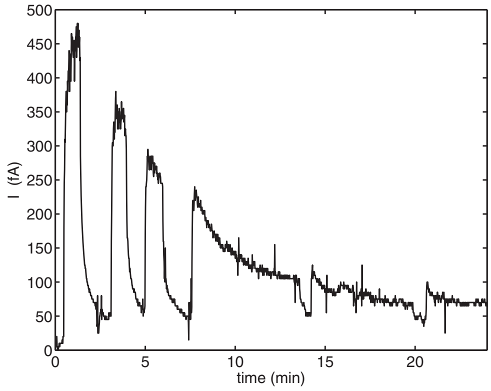

Fig. 2: Plot of the hyperconductivity current of a previously not irradiated crystal of fluorphlogopite when the alpha irradiation starts and is then interrupted for short intervals. The sample holds a reservoir of holes that has been created by the emission of electrons through the dominant β^- decay channel of ^{40}K during a long time. Quodons, created by alpha irradiation, trap and swept away the holes in this reservoir producing the large initial current peaks [2]. Thereafter the injection of holes comes only from the charge carried by the alpha irradiation. It also shows that the current continues to flow for a limited time in the absence of alpha irradiation. This is not discharge of the condenser formed by the crystal in the Faraday cage by a conduction current as no voltages are applied to the crystal. This is consistent with the persistence of quodons for several seconds in crystals of high quality and negligible iron content.

Results. – For practical application of hyperconductivity a material is needed that can be made synthetically under controlled conditions. A search identified phlogopite, fluorphlogopite, lepidolite and chrysotile as capable of supporting hyperconductivity, all of them phyllosilicates or layered silicates, and members with muscovite of the mica group, except for chrysotile from the serpentine group[3]. However, biotite, also from the mica group did not support hyperconductivity. Only fluorphlogopite of high quality can be synthesized commercially at atmospheric pressure; a sample had a volume resistivity of $1.2 \times 10^{17}\,\Omega$m. Other non-layered materials of high volume resistivity were tested for hyperconductivity. They were examined for possible use in contact with fluorphlogopite and muscovite. In the order of decreasing resistivity, they were PTFE, quartz, borosilicate glass and epoxy resin, none of which showed hyperconductivity. This suggests that the layered structure is a conducive condition for hyperconductivity but certainly not sufficient as the biotite example makes clear. The reason for that is presently unknown.

Persistence of current after alpha irradiation. This effect is seen in fig. 2, which shows the initial sweeping of holes from a crystal of fluorphlogopite. The plot shows that the build-up time to start the hypercurrent when irradiation commences is typically 1 to 2 seconds while the current persists for at least one minute.

This means that the quodon-current continues transporting charge for a long time after the end of alpha irradiation. With alpha-particle irradiation most quodons are created in atomic cascades. The energy of alpha particles from ^{241}Am is too low to cause induced alpha activity in the surrounding materials and excitation of electron shells yields insufficient momentum to create quodons. A foil of aluminium of 0.1 mm thickness inserted between the alpha source and the crystal stopped the creation of holes and quodons, which showed that the continued current flow was not due to X-rays from the decay of Am. When irradiation ceases, quodons continue to be created in cascades but molecular-dynamic studies have shown that this terminates in less than one picosecond [34]. The time for shutting the irradiation gate is less than 0.5 s. Modelling of breathers showed that they could be elastically reflected at a boundary and at sufficiently high energies they can eject the last atom when inelastically reflected [18]. Evidence from fossil tracks in muscovite that quodons could propagate up to 0.5 m at about 700 K showed that they could tolerate minor lattice defects. It is highly improbable that in propagating that far they did not encounter multiple dislocations. Such stability suggested that they might survive multiple reflections in a natural crystal. It would manifest as briefly continued flow of current after irradiation ceased as observed in the present experiment.

Figure 3 shows in more detail this prolonged decay in a crystal in which the charge reservoir had previously been depleted. Modelling studies of the speed of breathers in muscovite indicated that they are slightly subsonic of the order of a km/s [16] and the speeds of the other types of excitation are usually supersonic. The continued current flow after 50 seconds shows that some of the quodons must have travelled a distance in excess of 10 km at room temperature in a natural crystal. This suggests current flows both ballistically and by percolation. It explains why the positioning of terminals does not depend on crystal orientation. In contrast, fig. 4 shows the change of quodon-current following interruption of irradiation of a crystal of natural phlogopite of low quality with high iron content. There is no evidence for continued current flow. The only change in the measurement procedure was that of the crystals, thus excluding the possibility that the prolonged decay is an artefact of the equipment. The results reported here relate to very small currents. To see if this was a limiting factor the current density was increased in a sheet by reducing its cross-section area. After allowing time for annealing of possible crystal damage, a reduction of about 1000:1 in the cross-section area caused no change in the current with constant input conditions. The most likely cause for a limit is instability of quodons due to their close proximity. If they are stable when separated in a sheet by ten times the size of their envelope and by five times the spacing between layers, then an upper limit of order 10^5 A cm^{-2} might be possible.

[3]Chrysotile: this was suggested and provided by Prof. J. G. Fitton: it has a rolled-up structure.

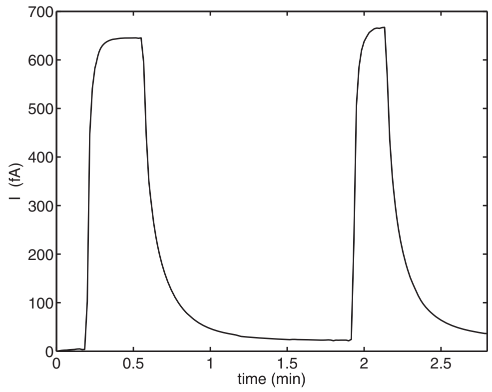

Fig. 3: Plot of the hyperconductivity current in a lepidolite crystal of good quality. In this case the reservoir of holes has been previously depleted by submitting it to alpha irradiation (see fig. 2). This plot shows the current when the alpha irradiation is started and interrupted twice, reaching similar peak values. Each time, after the irradiation is interrupted, the current still flows after 40 seconds. This is evidence of the delayed quodon current. This effect does not happen in fig. 4 because of the bad quality of the sample. The sample of lepidolite was 18 times thicker than the one used in fig. 2, which explains both the larger limiting current and the rounded profile.

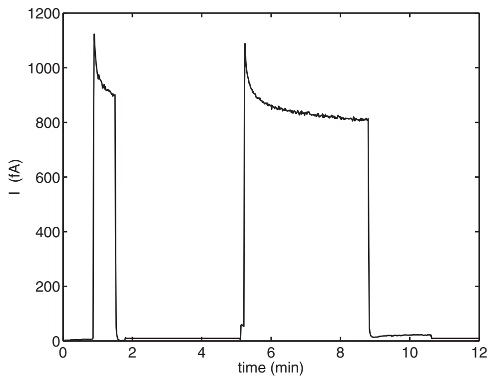

Fig. 4: Plot of the hyperconductivity current when the irradiation is interrupted. This is for a natural crystal of phlogopite of low quality with a high iron content. The crystal was black in colour, badly deformed with curving of the (001)-planes and fractures. There is no evidence of continuation of the current flow after irradiation is stopped.

Conduction and hyperconductivity currents. When irradiation begins the dominant effect is the creation of many quodons. Alpha particles can create many thousands of quodons but can introduce at most two units of charge. In a previously not irradiated crystal, there is a reservoir of holes created by the decay of ^{40}K. The dominant decay channel is β^- with 89% probability and leaves a positive charge at the decay site, where quodons are created, which can be carried by the resulting quodon to a remote site [10]. This creates a reservoir of holes near or on the potassium chains. The emitted electrons, however, are scattered away from the chains. When irradiation starts

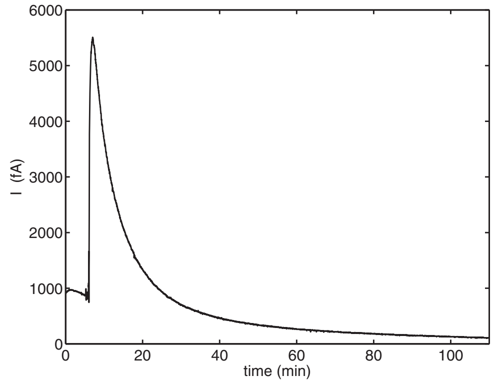

Fig. 5: Plot showing how the total current in muscovite decreases with time after irradiation begins. Initially it involves the depletion of charge stored in the crystal created by the beta decay of potassium. It tends to a limiting current from the alpha particles and any conduction current from an applied voltage potential. The rate of hole generation by K-decay is negligible. The sign of the current is conventional.

the resulting quodons can trap most of the holes but few of the electrons. The holes injected by the alphas progressively neutralise these electrons. This results in an initial surge of current, as shown in fig. 5 for muscovite, that decreases in time to a limiting current similar to the injected current. The influence of the quodons is illustrated by peak current of over 5000 fA during the sweeping-up of charge with only 40 fA input from the alpha source.

Three sources contribute to the limiting current.

Firstly, the residual charge in the hole reservoir, which is being replaced continuously but at a slow rate. Secondly, the holes injected by the alpha particles. Thirdly, a conduction current arising from any electric potential applied to the crystal. These can arise from contact potentials due to different metals at electrical connections to the crystal, a bias potential from the current amplifier or an external source. Figure 6 shows how these contributions meld into the observed current for a crystal of lepidolite.

The ability of the current amplifier to apply a bias potential to a crystal enabled the interaction between quodons and free charges to be examined. A bias of 5 V applied to a strip of fluorphlogopite caused a conduction current of 1.5 fA, giving a conductive resistance of $3.3\times10^{15}\,\Omega$. With end irradiation of the crystal strip of 0.8 mm thickness, only $\simeq 12\%$ of the available hole current of 40 fA, or 5 fA, was injected. There is a further reduction of about 24% in the current entering the strip due to the shorting of some of the current to the metal clamp holding the strip at the irradiated end, giving 4 fA input. Under these conditions the measured quodon-current was 300 ± 10 fA. To achieve this current by conduction would require a bias of 1000 V. This increase in the measured current in the presence of quodons is attributed to the much higher average speed of charge movement by quodon percolation relative to that of charge drifting in conduction. Similar

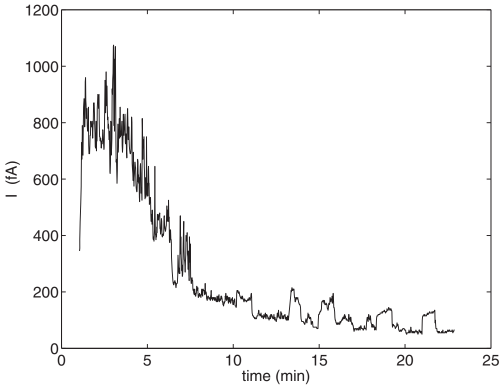

Fig. 6: Plot showing how the current varies after the creation of quodons is briefly interrupted for a crystal of lepidolite. Initially the current stems from a charge reservoir. As this is exhausted the contribution from the pulsed injection of holes emerges, causing the bottom part of the pulsed current. The upper part of the pulse occurs when there is no injection of holes, leaving only the conduction contribution. Contact and bias potentials associated with the current amplifier cause the conduction current.

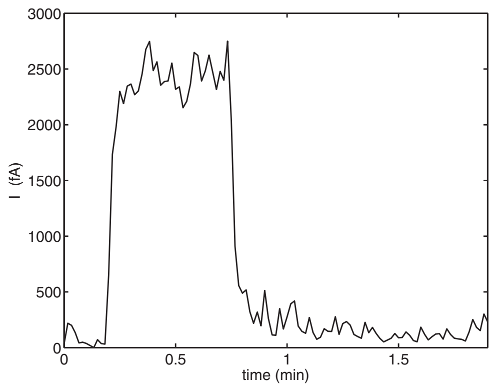

Fig. 7: Plot of the current as alpha-particle irradiation of crystal is pulsed on then off with 5 V bias potential applied to a crystal of fluorphlogopite. It shows how quodons increase the current by increasing the movement of charge.

results were obtained when the bias potential was zero but a potential was applied to the irradiated end of the strip. A plot of the change in current is shown in fig. 7 for fluorphlogopite.

Annealing by hyperconductivity currents. Crystals of muscovite and similar micas are easily cleaved in the (001)-plane but present difficulties in machining in other directions. To study propagation of charge in this plane when using a weak source of alpha particles, it was thought necessary to use crystals with excellent natural faces. As these were difficult to obtain a study was made of the performance of machined surfaces. It was found that the current flowing through a crystal from a machined surface progressively increased when irradiated with alpha particles, indicative of annealing of defects by quodons [33,35]. Since the range of 5 MeV alphas in mica is about 3 μm

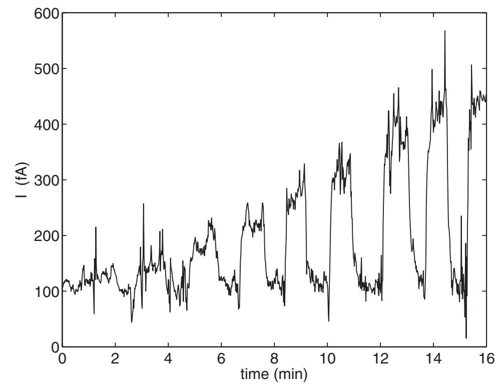

Fig. 8: Plot showing the growth of the transmitted hole current as crystal damage is annealed in a crystal of muscovite. An edge of a strip of crystal was machined and then exposed to alpha particles. Although the alpha particles caused further damage to the lattice locally near the surface, the influx of many quodons progressively annealed damage throughout the crystal. Due to the stability and internal reflection of quodons they are present in all parts of the crystal. The flux of alphas was pulsed. This enabled a study of the rate of the fall of the current following interruption of the injected alphas.

only quodons could affect the majority of the damaged zone of the crystal that extended to more than 50 μm. It was found that the transmitted current went from near zero to about that typical of a natural face after 20 minutes of irradiation. This amounted to at least 10^{10} quodons passing into the damage zone of approximate volume 2×10^{-3} cm^3. Almost any handling of crystals, such as applying electrical contacts or mechanically clamping, degraded their performance but it could be restored by quodon annealing. This is illustrated in fig. 8 for muscovite. It does not matter where quodons are created as they rapidly disperse through a crystal.

Hyperconductivity current and interfaces. Crystals with the required structure do occur naturally, such as lepidolite, but usually contain impurities and structural defects. Fluorphlogopite of high purity can be grown artificially and looks suitable for practical applications; it is grown as boules of relatively short length. The layered structure enables sheets to be cleaved from which strips can be cut. For practical use it would be useful to join strips. It seems impossible to prepare the ends of strips to get structural alignment and contact at the atomic level. Two simple ways to join strips of crystal or fluorphlogopite were examined: overlapping and butt joining of tapered ends. In both cases hyperconductivity currents were found to propagate through the interface with modest efficiencies between 12% and 23% but only after a period of annealing by the quodons. Figure 9 shows how annealing of the damaged interface allows quodon-currents to pass through. Surrounding a strip with an epoxy resin or glass caused no measurable decrease in the measured current.

The perceived stability of quodons depends on the quality and composition of layered crystals. From the point of

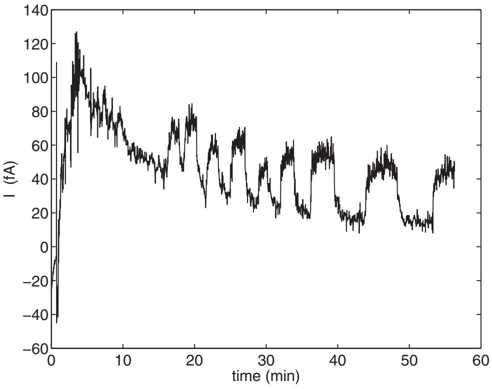

Fig. 9: Plot of the hyperconductivity current passing across an interface between two strips of a crystal of fluorphlogopite. The surfaces were cleaned mechanically, pushed together and held by an insulated clamp. This would have caused irregular and damaged interface surfaces with gross misalignment of the lattice near the interface. Contact would be erratic at the rough surfaces. Despite these obvious defects it was found that quodons annealed defects at the interface to allow current to flow. The efficacy of quodon annealing is high. In a damaged region of volume 1 mm^3 containing about 10^{21} atoms, the annealing was completed by about 10^{10} quodons in about twenty minutes.

view of the theoretical description of quodons, it is hard to see how they could propagate along misaligned chains in a potassium sheet and impossible to migrate from one potassium layer to an adjacent one. The positrons emitted in the decay of ^{40}K are diffraction scattered by the lattice and the quodon created by the recoiling nucleus has approximately the same de Broglie wavelength so they might quantum tunnel. Despite these uncertainties, the described experiments seem to demonstrate that quodons can travel through interfaces and progressively repair the defects encountered.

Persistence of hyperconductivity under magnetic fields. Large crystals of muscovite often contain ribbons of black magnetite. Although they appear extensive, they seldom exceed 0.2% by mass of the total. However, magnetite is electrically conducting, which makes them unsuitable for most electrical applications. Magnetite also is magnetic, which could interfere with magnetic fields. Synthetic fluorphlogopite is free of magnetite and so is suitable for applications involving strong magnetic fields. It is likely that neutral quodons are not influenced by magnetic fields but the situation for quodons with charge was not known and so it was examined. The most sensitive test would be on the survival of charged quodons in a magnetic field. This was examined by measuring the rate of decay of the quodon-current following interruption of irradiation in fields of zero, 0.5 and 1.1 tesla. Within measurement error it was found to be independent of the field strength.

Discussion. – Hyperconductivity experiments show that charge transport in the absence of an electric field can be produced when a crystal is bombarded by alpha particles that produce a big number of quodons and, therefore, a current large enough to be measured. However, the fossil tracks of quodons with a positive charge recorded in muscovite enable the properties of individual quodons to be studied. The recording process ceases to operate below approximately 700 K [36]. The track shown in ref. [10] is 29 cm long and has generated 50 secondary quodons that are strong enough to hold a positive charge. The long range of quodons measured in the ejection experiment at 300 K [1], when the recording process cannot operate, shows they do not gain energy from the precipitation process. The total energy of the multiple secondaries stems from the primary, indicating an energy range of at least 50:1 for stability and holding a charge. The secondaries frequently create further secondaries, thus increasing the range of energies for quodons to be stable. Assuming a plausible initial energy of 50 eV [10] for the primary suggests a possible lower energy for stability of tenths of eV. It would be instructive to explore the use of a plasma source with energies of a few eV to test this hypothesis.

The mobility of charge in quasi–two-dimensional systems has attracted much attention [13,37,38]. This is partly because high-T_c superconducting (HTSC) materials have a layered structure. This superficial similarity to muscovite prompted a study to see if the cuprate layer in some HTSC materials might allow quodons to exist [39]. Subsequent numerical modelling showed that breathers could exist in a flat CuO plane [40]. It is possible that breathers could be created naturally at low temperatures by stochastic thermal processes [41–43] but it is not known if that is possible for highly mobile quodons. In any case, a major difference between HTSC and HC is that HC involves only single charges. The reason for the layered structure of HTSC materials remains intriguing. For HC materials the layered structure and mixture of atoms of different masses create the conditions necessary for the existence and stability of quodons. Kink-like lattice excitations have been proposed for the cause of fan-shaped fossil tracks in muscovite, which show lateral instability [44]. The observed propagation of the excitation responsible for fans for up to 12 cm length requires that the excitation gains energy from the recording process to compensate for the lateral spread.

Conclusions. – We have extended previous studies of hyperconductivity in muscovite to other layered silicates, namely, lepidolite, phlogopite, chrysolite and synthetic fluorphlogopite but it was not observed in biotite. The presence of iron in the crystal structure impedes hyperconductivity. Nor was it found in the construction materials PTFE, quartz, borosilicate glass and epoxy resin. We have found that hyperconductivity is insensitive to minor crystal defect and can even anneal some of these. It enhances conduction current by two orders of magnitude and it is not affected by magnetic fields up to 1.1 T. These results imply striking properties of stability, persistence, and long flight-paths of charge-carrying, nonlinear excitations. We think that the present work makes significant advances on the phenomenon of hyperconductivity with potential for technological applications.

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