



## Rapid Communication

## Sunlight-activated near-infrared phosphorescence as a viable means of latent fingerprint visualisation



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## ARTICLE INFO

## Article history:

Received 16 January 2017

Received in revised form 18 April 2017

Accepted 18 April 2017

Available online 26 April 2017

## Keywords:

Infrared

Fluorescence

Phosphorescence

Fingerprint

Powder

Time-resolved

## ABSTRACT

The chromium-doped zinc gallogermanate,  $Zn_3Ga_2Ge_2O_{10}:0.5\%Cr^{3+}$ , was prepared and modified for use as a near-infrared (NIR) luminescent fingerprint dusting powder. The optical properties of this material are illustrated and have been shown to offer versatility and promise in terms of overcoming troublesome backgrounds during latent fingerprint examination and imaging. The highly fluorescent NIR emission of the powder allows inherently fluorescent and patterned substrate backgrounds to be mitigated, whilst the phosphorescent nature of the material allows all conceivable background interference to be removed, whilst also constituting the first reported use of a NIR phosphor for latent fingerprint enhancement and imaging.

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## 1. Introduction

Latent fingerprint enhancement has remained a pivotal tool within forensic science casework for over 100 years, serving as a fundamental and crucial method of attributing contact between an individual and an item of evidential value [1–3]. Despite the relative maturity of the discipline and the multitude of visualisation processes that have been documented and implemented over the years, significant challenges remain within the fingerprint enhancement community. The evolution of novel material types and compositions that are used during the manufacture of commonly encountered everyday items (recycled plastics, for example) or technologically driven substrate changes (3D printed items) means that the forensic practitioner has to re-evaluate, assess and validate pre-existing processes for their suitability on such evidence types, whilst also continuing to explore more effective and pertinent processes specific to the medium in question.

Process selection aside, a fundamental long-standing issue that is frequently encountered during sample examination, is the inability to remove background detail that often obscures some, or all, of the developed fingerprint that is to be imaged [4,5]. Such issues often arise from complicated patterned and/or multi-

coloured backgrounds that make fingerprint enhancement process selection difficult due to a lack of colour contrast or differentiation between developing agent and the background on which the fingerprint resides [6], or from substrate backgrounds that are composed of fluorescent components (inks, pigments, embedded security markings) that interfere with fluorescent fingerprint enhancement processes and subsequent visualisation in the visible spectrum [4,5].

Several years ago, Ma et al. reported the use of anti-stokes up-conversion powders as an amenable process that went some way into overcoming background interference issues [7], given the fact that few anti-stokes fluorescent features are found on conventional non-secure items (items such as passports, currency, identity cards, and security labelling often have anti-stokes features embedded within them, however). The use of an anti-stokes treatment and imaging approach [7,8], requires the use of high intensity invisible IR laser excitation that may be destructive to certain evidence types (thin films and plastics, for example) as well as being restricted to laboratory use given the excitation sources and safety requirements needed during processing. Moreover, because the fluorescent output of the anti-stokes powders tend to fall in the visible spectrum (green is the most common), background substrate artefacts still remain in the resulting 'fluorescent' image and may still cause interference if the developed fingerprint is weak and/or if the background retains some of the powder that has been applied to it.

Menzel [9–11], amongst others [12–15], reported the use of visible phosphorescent materials as a method of mitigating some

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background hindrance on various substrates. At the time, commercially available imaging and illumination technology that readily facilitated such an approach was in its infancy, and the setup designed by Menzel, although effective, was cumbersome and not readily transferable to mainstream forensic laboratories [12]. This work did, however, illustrate the usefulness of phosphorescent imaging, whereby a suitable phosphor (typically a lanthanide (doped) complex [11]) may be applied directly to the target substrate (either as a cyanoacrylate dye stain or as a standalone process) and the fluorescent response stimulated with a low wavelength excitation source (UV to blue). In instances where the background did not fluoresce under the same excitation wavebands, or where the existing design does not impede on the fluorescent imaging, standard fluorescence examination was suitable. On occasions where interference did occur, then phosphorescent imaging could be exploited, whereby the illumination source is turned off and the short-lived phosphorescence (“after-glow”) of the developing reagent may be observed *via* time-gated imaging. Visible artefacts often remained in the resulting images, but the contrast between fingerprint and background was significantly better than standard fluorescence approaches. To date however, there has been little further development, or research, into the use of phosphorescent reagents and imaging for latent fingerprint enhancement.

Recently, novel fingerprint enhancement approaches have been reported that further explore the notion of interfering background mitigation through fluorescence at extended wavelengths in the near-infrared (NIR) [16–19]. Imaging most substrates in the NIR (700–1100 nm) often facilitates the muting of many background patterns and textures, given the fact that these designs do not transmit at higher wavelengths. Most visible patterns that we are used to seeing are designed to have maximum contrast in the visible spectrum, that is 400–700 nm. The development of NIR fluorescent reagents, principally in the form of fingerprint dusting powders, by Chadwick et al. [16] and King et al. [17], illustrated the capacity of visible wavelength induced NIR fluorescence as a means of quenching background interference across many pertinent substrates (drink containers, wrapping papers, polymer currency, etc.), whilst also ensuring that the mode of fingerprint enhancement was as simple as possible. The non-toxic nature of the algae based powder reported by King is a useful property to consider when exploring novel reagents for use *en masse* in both the laboratory as well as at the crime scene [17]. King et al. recently reported further developments relating to NIR fluorescence for fingerprint visualisation by proposing the use of finely milled cuprorivaite powder, a base mineral found in the ancient pigment, Egyptian Blue [18]. The fundamental aspects exploited within this work showed that both the excitation wavelength and intense fluorescent emission of cuprorivaite could be readily achieved within the NIR (excitation: 730–800 nm, emission: 850–1050 nm), illustrating the first example of a NIR-NIR fluorophore for use as a fingerprint dusting powder. The extended excitation and emission wavelengths used in this study were able to overcome nearly all conceivable background interference/artefacts that may otherwise arise under visible or visible-induced fluorescence examination. The versatility of cuprorivaite was acknowledged by Errington et al. who also reported its promise as a fingerprint dusting powder, this time using visible wavelengths (white or red) to induce the NIR fluorescence [19].

The evolution of novel materials within all aspects of materials science should be of particular interest to the forensic community given its multi-discipline remit and ever-continuing need for the development of new reagents, technology and infrastructure to enhance the forensic workflow. In 2011, Pan et al. reported the synthesis of a Cr<sup>3+</sup>-doped zinc gallogermanate that has long-persistence NIR luminescence under sunlight activation [20]. This

NIR persistent material was considered to have potential applications that included night-vision surveillance, solar energy utilisation and *in vivo* bio-imaging. The strong NIR emission between 650–1000 nm was readily achieved following short (seconds to minutes) sunlight activation with an afterglow that extended, in some cases, to 360 h. Pressed discs, bulk powder and oil-based paints of Zn<sub>3</sub>Ga<sub>2</sub>Ge<sub>2</sub>O<sub>10</sub>:0.5%Cr<sup>3+</sup> were all shown to possess excellent NIR phosphor activity, with long-term stability [20].

Herein, we report the use of a tailored Zn<sub>3</sub>Ga<sub>2</sub>Ge<sub>2</sub>O<sub>10</sub>:0.5%Cr<sup>3+</sup> powder, as a (sunlight activated) NIR phosphor for the detection and visualisation of latent fingerprints from a range of notoriously troublesome substrates. For the first time, the benefits of *both* NIR luminescence and NIR phosphorescence for fingerprint visualisation are explored with the ability to completely overcome and remove background complexities and interference that would otherwise prove problematic in terms of imaging.

## 2. Methods

Zn<sub>3</sub>Ga<sub>2</sub>Ge<sub>2</sub>O<sub>10</sub>:0.5%Cr<sup>3+</sup> was prepared in a manner analogous to that reported by Pan et al. [20]. ZnO (≥99.99%), Ga<sub>2</sub>O<sub>3</sub> (≥99.99%), GeO<sub>2</sub> (≥99.99%) and Cr<sub>2</sub>O<sub>3</sub> (≥98%) (all purchased from Sigma Aldrich, UK) were ground, in stoichiometric amounts, to a fine powder, using an agate pestle and mortar. For a typical synthesis, Ga<sub>2</sub>O<sub>3</sub> (3.000 g), GeO<sub>2</sub> (3.350 g), ZnO (3.908 g) and Cr<sub>2</sub>O<sub>3</sub> (0.513 g) were used. The mixture was pre-fired at 900 °C for 2 h, in an Elite Thermal Systems furnace oven, before being allowed to cool to room temperature and re-ground using an agate pestle and mortar. The material was then sintered at 1150 °C for 2 h, before being allowed to cool. At this stage the resulting solid was extremely hard and required forceful grinding with an agate pestle and mortar to obtain a true powder. This powder was placed into a Fritsch Pulverisette planetary ball mill and loaded with zirconia beads (Zirbeads™, 1.4–3.35 mm diameter) as the grinding medium. Following 10 min of grinding, a fine, free-flowing Zn<sub>3</sub>Ga<sub>2</sub>Ge<sub>2</sub>O<sub>10</sub>:0.5%Cr<sup>3+</sup> powder was obtained (*ca.* 10.5 g), with a mean particle size of approximately 10 μm, as determined through use of a Malvern Masterseizer 2000 Particle Size Analyser.

All fingerprints deposited during these investigations were taken from a range of 5 donors (1 female, 4 male, all aged 25–50 years) at varying times across the day so as to create a ‘natural’ working portfolio of latent deposits. Donors were asked not to wash their hands during the 30 min preceding mark deposition, thereby ensuring natural secretions were present on the papillary ridge skin in a manner representative of operational samples. Donors were asked to rub their hands together prior to mark deposition to ensure that all residues and any environmental contaminants were evenly distributed across all fingers. For the purpose of this preliminary study, single fingerprints were deposited by each donor on a range of substrate types. Approximately 5–10 samples of each substrate type were processed. Latent fingerprints were typically aged for 1–7 days prior to development.

A range of substrates were selected for this study that were representative of evidence types that are commonly considered difficult to image due to inherent complexities within their composition. Typical examples included: polished and patterned metal surfaces (stainless steel, aerosol cans, drinks cans, copper), polymer banknotes, mirrored surfaces and the data side of compact discs (CDs).

Latent fingerprints were developed using a squirrel hair brush (BVDA). Zn<sub>3</sub>Ga<sub>2</sub>Ge<sub>2</sub>O<sub>10</sub>:0.5%Cr<sup>3+</sup> powder was loaded onto the brush fibres and gently passed over the substrates to reveal fingerprints. Any excess powder was easily removed by passing a clean squirrel hair brush back over the surface.

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