Dear Editor,

we thank you for your time and consideration, and we thank the reviewers for their very useful and supportive comments.

We revised our manuscript in order to address the critical remarks they highlighted with the purpose of further strengthening the paper.

A detailed list of our revisions is reported in the following, and the corresponding changes are highlighted in red in the marked-up version of the manuscript, that we have also carefully checked and revised.

We believe that with these modifications and integrations we have addressed the comments raised by the Reviewers, therefore we re-submit the new version of the paper for your and their consideration.

Looking forward for your feedback, best regards

On behalf of the co-authors,



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Reviewer #1

In this review, the authors give an overview over the current state-of-the-art in research on color centers in a wide range of materials as possible single photon sources. The review is very timely, it is well written and I recommend publication after some minor issues have been addressed:

1.1 - p. 1 l. 55: There seems to be something missing in "with significant subsequent leading to their practical implementation"

We apologize for the truncation. We addressed this issue by modifying the text as follows:

> Single-photon sources, i.e. physical systems offering the *on-demand* emission of individual photons with desired properties, are key ingredients for current and prospective applications in integrated photonics and quantum technologies [Eisaman11]. The concept of single photon sources originated in the late 20th century, with a significant subsequent body of work leading to their practical implementation.

1.2 - p. 1 l. 86: What does "most promising materials" mean, i.e. most promising according to which criterion?

We expanded this point in the revised version as follows:

- In the abstract:

  > This review discusses the emerging material platforms hosting single-photon emitting color centers, with an emphasis on their potential for the development of integrated optical circuits for quantum photonics.

- in the Introduction section::

  > Single-photon sources, i.e. physical systems offering the *on-demand* emission of individual photons with desired properties, are key ingredients for current and prospective applications in integrated photonics and quantum technologies [Eisaman11]. In particular, the relevant properties that a single-photon source must have to enable the realization of these technologies generally include the photostable and on-demand delivery of fully polarized single indistinguishable photons. Besides, the ideal single-photon source is a scalable physical system operating at room temperature and characterized by a high emission count rate and quantum efficiency.

- in the “Perspectives” section, we added on the trade-offs required for specific quantum-enhanced technologies:

  > The quest for optimal classes of solid-state color centers and their integration in integrated quantum photonics platforms is a buzzing research field which has achieved impressive developments in the last decade. Several quantum systems have been demonstrated in different materials, each offering peculiar properties and advantages combined with specific limitations. From the point of view of the maturity of each considered material as a platform for integrated quantum photonics, different challenges are still to be addressed. However, depending on the specific quantum protocol to be implemented, the availability of color centers with different emission

properties allow one to identify a suitable trade-off between the manufacturing opportunities and the accompanying drawbacks. For instance, quantum-enhanced sensing protocols require the optical addressability of the spin system and its coherent manipulation, without the strict need for high-indistinguishability or NIR/IR emission wavelengths. Conversely, the development of quantum-integrated photonic circuits requires a hyperfine coupling with nearby nuclear spins as a key parameter for transferring quantum information between the electron spin of the quantum emitters and the proximate nu-clear spins of the crystal environment. Furthermore, the realization of solid-state quantum memories does not necessarily find its limitation in the VIS emission wavelengths when long distance communication is not demanded. If the realization of larger programmable photonic processors is targeted [XXue21], i.e. characterized by a large number of qubits and thus capable of processing higher complexity algorithms, industry-scale-compatible manufacturing processes could be beneficial together with a consolidated maturity of the material platform synthesis (e.g. silicon CMOS technology). Moreover, if the emitted single photons are intended to be used in quantum processing systems based on commercial optical fibers, the emission in the telecom band represents an important prerequisite to minimize the losses. On the other hand, indistinguishable photons constitute the prerequisite for quantum interference effects underpinning the linear quantum optics paradigm [Aharonovich16, Aaronson11]; consequently the availability of solid-state single photon emitters with Fourier transform limited emission lines is crucial for this range of applications.

We also added a set of tables summarizing the emission properties of color centers related to each class of emitters.

1.3 - p. 4 The abbreviation "HBT" used in the figure caption should be properly introduced in the main text.

Thank you for spotting this inconsistency. We replaced the acronym HBT with the full name of the technique in Fig. 1 Caption:

> **Figure 1.** Color centers and experimental characterization. **a)** Crystallografic structure and **b)** fine level structure of group IV color ceters in diamond [Iwasaki20]. **c)** Single-photon sensitive confocal microscope connected to a Hanbury-Brown&Twiss interferometer [GattoMonticone14]. **d)** Example of a second-order auto-correlation function $g^2(\tau)$ measurement for a single MgV color center in diamond [Corte23]. **e)** Typical background-substracted emission rate curves as a function of the optical excitation power for the saturation intensity estimation according to the saturation model [Corte23].

Reviewer #2

The manuscript represents one of the first attempts at a review that describes solid state color centers for single-photon generation in several different materials. The text of the manuscript is based on current articles up to 2023 publication and correctly describes the main trends in the research of solid state color centers for single-photon generation. However, in my opinion, this manuscript needs to be improved.

2.1 - The figures are excessively small in size and all of them are placed in the text 1-2 pages later than they are referenced in the text. The drawings themselves are practically not discussed in the text of the review.

We performed the following revisions:

- We modified the arrangement of Fig. 2-5 to enhance their readability. In the process, we removed some figure subsets (Fig. 3, Fig. 5) to allow for larger magnification of the remaining ones.
- We referenced the figures in the main text as suggested.
- We also modified the position of the figures in the text (they were simply placed at the end of the paragraph of interest) in a more appropriate position.

2.2 - The manuscript contains virtually no description of desired properties of solid state color centers for single-photon generation. It would be advisable to state them clearly at the beginning of the manuscript.

This point has been expanded as suggested by both Reviewers. A detailed description of the changes has been given in reply to Comment 1.2.

2.3 - The manuscript does not contain Tables containing information on the parameters of solid state color centers for single-photon generation in the materials discussed. Such tables are traditionally present in other similar reviews and greatly facilitate understanding of the general state of affairs in this area of research.

We included a summary table at the end of each section dedicated to materials platforms:

		ZPL (nm)	T (K)	Excited state lifetime (ns)	FWHM at operational temperature (nm)	Single emitter saturation count rate (kcps)	Spin manipulation	Integration of SPs with photonic structures	References
Diamond	NV	575, 638	RT	12-22	Broad emission	1 k	Yes	Yes	[16], [30]-[34], [58], [62]
	SiV	738	RT	1.0-2.4	0.7 – 5	~4.8k	Yes	Yes	[35], [57], [59]-[61], [63], [66]
	GeV	602	RT	1.4-5.5	5	170-1.2 k	Yes	Yes	[35], [37], [42], [53], [66]
	SnV	620	RT	6	6	530	Yes	Yes	[35], [38], [64], [65]
	PbV	520, 552	RT	>3	7	1.04 k			[35], [39]
	MgV	558	RT	2.4	3	0.44-1.46 k			[27]
	ST1	557	RT	9		~5	Yes		[45]
	He	536, 560	RT	29 , 106	<2 (ensemble)				[46]
	Xe	794, 812	RT	~0.77	Broad emission				[47]

		ZPL (nm)	T (K)	Excited state lifetime (ns)	FWHM at operational temperature (nm)	Single emitter saturation count rate (kcps)	Spin manipulation	Integration of SPs with photonic structures	References
Silicon Carbide	V_{Si}	858.2, 862, 917	RT	~5	<100	~10	Yes	Yes	[71]-[76], [80], [94], [96]
	$C_{Si}V_C$	650 – 675	RT	1.2	>100]	2 k			[71], [80], [83]
	NV	1150 – 1350	RT	2	1-3	17.4	Yes		[90], [91]
	V4+	1279-1387	RT	>70			Yes		[92]

		ZPL (nm)	T (K)	Excited state lifetime (ns)	FWHM at operational temperature (nm)	Single emitter saturation count rate (kcps)	Spin manipulation	Integration of SPs with photonic structures	References
Silicon	G	1279	4-110K	35.8	0.28	8	Yes	Yes	[103], [107], [127], [100], [140]
	W	1218	4-60K	3-34.5	0.8	2-6		Yes	[108], [114], [138],[139]
	T	1326	<5 K	940	0.01	2	Yes		[112], [113]

		ZPL (nm)	T (K)	Excited state lifetime (ns)	FWHM at operational temperature (nm)	Single emitter saturation count rate (kcps)	Spin manipulation	Integration of SPs with photonic structures	References
(Mg)-doped and undoped GaN films	Intrinsic defects	600-700	RT	~1-3	~5	100-150		Yes	[150], [151]-[153]
	Intrinsic defects	1100-1300	RT	0.7	3-50	500		Yes	[155],[156]
AlN	$N_{Al}V_N$, $V_{Al}V_N$	550-1000	RT	~2	<12	500		Yes	[159], [160], [166]
hBN	V_NN_B	623	RT	~3	~3	>4 k		Yes (without attribution to a specific defect complex)	[167]-[169], [175]-[178]
	V_BC_N	630	RT	~2-6	~5-35	>4 k			[167], [172], [175]-[178], [188]-[195]
	V_B^-	850	RT	1.2	Broad emission	>4 k	Yes		[171],[173], [174]-[178]
SiN	Intrinsic defects	567-670	RT	3.8	Broad emission	500		Yes	[205]

2.4 - The manuscript is overly laconic. For example, only half a page of text is devoted to the properties of color centers for single-photon generation in hexagonal boron nitride. At the same time, the volume of similar reviews on this material (DOI 10.1088/1361-6633/ab6310; https://opg.optica.org/col/abstract.cfm?URI=col-20-3-032701) amounts to 20 pages of text and more. It would be correct to either supplement the text of the manuscript or at least list these reviews at the References.

The scientific research on hBN color centers is indeed flourishing, as it is for the other classes of materials discussed in this review, as they find ample work in the scientific literature. The reasoning behind our choice is to give a space to SPSs in hBN proportional to the amount of classes of emitters currently identified.

We included the suggested references as follows:

> The localized fabrication of emitters in hBN has achieved so far with a variety of experimental methods [Huang22], including thermal annealing [Tran16_2], plasma treatment [Xiao22], femtosecond pulsed laser [Songyan17, Gao21], electron [Tran16_2, Fournier21] and particle irradiations [Gu21, Guo22], and strain field engineering [Grosso17, Proscia18]. These results, in parallel with significant efforts to shed light on the chemical nature of the discussed classes of emitters [Sajid20], indicate the readiness of the material for the scalable fabrication of arrays of quantum emitters.

2.5 - There is no mention in the manuscript of highly selective formation of color centers for single-photon generation in the volume of diamond by intense laser radiation.

We included a dedicated discussion in the section devoted to diamond color centers

Moreover, diamond color centers can be straightforwardly fabricated by means of the incorporation of selected impurities by ion implantation due to the relative simplicity of their defective structure, although their formation efficiency heavily depends on post-implantation activation processes [Luhmann21, Wang21, Gorlitz22, Chen17, Nieto23]. Furthermore, laser writing has recently disclosed a novel approach in diamond defects engineering for device fabrication, unlocking in-plane high spatial accuracy in the controlled placement of single emitters [Chen17][Chen19] and new perspectives for their integration in diamond chips [Sotillo16]. With respect to the doping of a diamond substrate with the desired atomic species during the chemical vapor deposition or high-pressure high-temperature synthesis of the material [Balasubramanian20, Ekimov18, Palyanov15], ion implantation, possibly combined with high-power laser processing [Engel23 ,Wang22] has the advantage of delivering a controlled number of impurities at specific locations in the target crystal, with spatial accuracies below 100 nm **(Fig. 2b)** [Schroder17, Schukraft16].

2.6 - The review also does not reflect numerous publications on single-photon electroluminescence in diamond (see for example, 10.1063/1.3670332; 10.1038/nphoton.2012.75; 10.1002/qute.202100048).

We agree to expand the amount of citations related to diamond electrolunminescence with respect to what already reported in the review. In addition to the 2012 work by Mizuochi, already included in the original manuscript, we inserted the following references:

[Lohrmann11] A. Lohrmann et al., Diamond based light-emitting diode for visible single-photon emission at room temperature, Appl. Phys. Lett. 2011, 99, 251106, 10.1063/1.3670332

[Forneris15] J. Forneris et al., Electrical stimulation of non-classical photon emission from diamond color centers by means of sub-superficial graphitic electrodes, Sci. Rep. 2015, 5, 15901. 10.1038/srep15901

Conversely, we hesitate to include the suggested work authored by D. Fediyanin since its diamond-related part relies mostly on the aforementioned papers, but we are open to do so upon the feedback of the Reviewer.

The main text was modified as follows to reflect these changes:

Furthermore, despite some noteworty achievements [Lohrmann11, Mizuochi12, Forneris15, Bray20] the electrically insulating properties of diamond challenges the electrical excitation of single-photon emission from individual color centers, which would boost the integration of these systems in high-density opto-electronic devices.