Single-Photon Emitters in Aluminum Nitride by Zr ion Implantation

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Abstract: We report on the generation of single-photon emitters in aluminum nitride films through Zr-ion implantation, which was predicted to form optically addressable spin defects. We studied implantation conditions, post-implantation procedures, and properties of resulting emitters.

Aluminum nitride (AlN) is a promising material platform for integrated quantum photonic technologies. First-principles calculations have shown that transition-metal impurities, such as Ti and Zr, can form stable complexes with nitrogen vacancies resulting in defects possessing a high-spin ground state [1] [2]. Recently, it was shown that Zr implantation into AlN leads to emission in the visible range due to Zr-vacancy defects [3]. However, the single-photon properties of this emission were not studied. Single-photon emitters (SPEs) were recently found experimentally in AlN films grown by metal-organic chemical vapor deposition, as well as in commercially available AlN substrates without any additional treatment of the samples [4–6]. Also, He ion implantation was applied to generate emitters in AlN samples, though it was found that implantation by itself increased the overall AlN photoluminescence but did not increase the detectable emitter density [7].

In this work, we studied the generation of single-photon emitters in AlN through implantation of Zr ions, which were theoretically predicted to exhibit spin properties. In the initial study, we used a broad beam ion implantation to determine the implantation energy, fluence, and post-implantation procedures required to generate single-photon emitters. Specifically, we prepared a series of samples using a different Zr ion implantation fluences Φ_{Zr} to study their impact on emitter formation and employed thermal annealing to activate emitters.

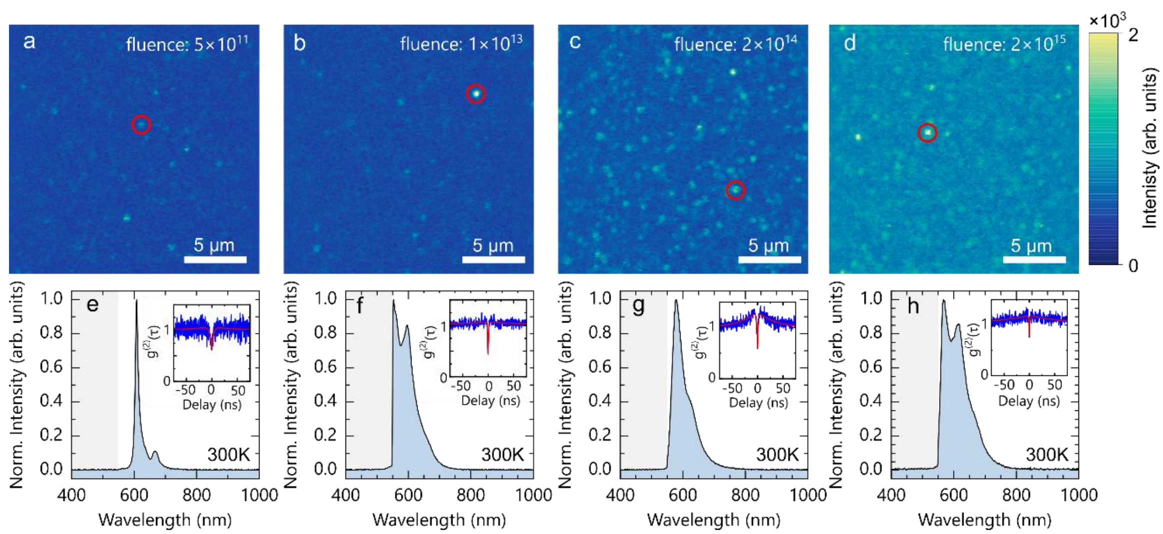

Figure 1. Density of single-photon emitters in AlN as a function of Zr implantation fluence Φ_{Zr}. Room-temperature photoluminescence (PL) intensity maps of samples prepared using Φ_{Zr}: (a) 5×10^{11}, (b) 1×10^{13}, (c) 2×10^{14}, (d) 2×10^{15} cm^{-2}. (e-h) Room-temperature photoluminescence spectra of emitters identified in PL maps (a-d), respectively. The PL data were background-corrected and normalized to the maximum intensity. Insets: second-order autocorrelation histograms recorded for the same emitters.

Photoluminescence (PL) intensity maps of the resultant samples revealed an increase in the density of isolated emitters with greater implantation fluence (Fig. 1a-d). We found that the AlN background fluorescence also increases as a function of Φ_{Zr}. The optimal ratio between the emitters density and the background fluorescence induced by ion implantation was obtained for $\Phi_{Zr} = 2 \times 10^{14}$ cm^{-2} (Fig. 1c). The single-photon nature of these emitters was confirmed by performing the second-order autocorrelation $g^{(2)}(\tau)$ measurements. We observed moderate values of $g^{(2)}$ at zero delay time, in the range of 0.2-0.6. These values can be considerably improved by spectral filtering and background correction. The structure of photoluminescence spectra of individual emitters resembles a zero-phonon line (ZPL) accompanied by lower energy phonon side-bands (PSBs) (Fig. 1e-h). The room-temperature ZPL linewidth varies between different emitters, which may be related to different implantation conditions, but will require further analysis to establish a correlation. At liquid-nitrogen temperature (77K), single-photon emitters with substantially narrower of ZPL linewidths were identified. Moreover, the emission was found to predominantly occur through the ZPL transitions. We estimated the Debye-Waller factor to be about 22, 43, and 62% for PL spectra of SPE_1, SPE_2, and SPE_3 emitters, respectively (Fig. 2a).

Presently, the ZPL peaks were observed at longer wavelengths than theoretically predicted for Zr-related defect complexes. We attribute the observed single-photon emitters to defects introduced by implantation damage of the AlN lattice, which may not contain Zr species as a part of defect complexes. To understand the role of Zr, we prepared another series of AlN samples implanted with Kr ions and will report on the comparative study. The implantation conditions with Kr ions were selected in a way to provide the similar damage as by Zr ions, but without any chemical doping effect.

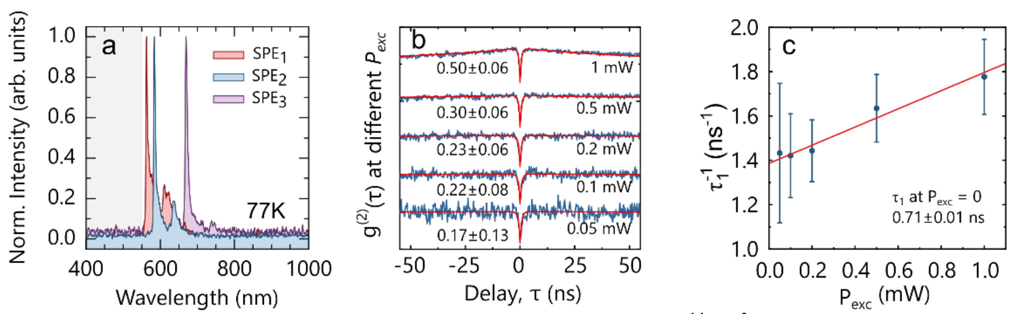

Figure 2. Characterization of single-photon emitters from an AlN sample with $\Phi_{Zr} = 2 \times 10^{14}$ cm^{-2}. (a) Photoluminescence spectra of different emitters recorded at 77K. The PL data were background-corrected and normalized to the maximum intensity. (b) Power-dependent second-order autocorrelation histograms of a representative emitter. (b) Power-dependent reciprocal value of τ_1, extracted from $g^{(2)}(\tau)$ fitting, characterizing the transition between the ground and excited state. A linear fit reveals the lifetime of the excited state of 0.71±0.01 ns.

We also performed excitation-power-dependent measurements of the second-order autocorrelation histograms of a representative emitter from the sample implanted with $\Phi_{Zr} = 2 \times 10^{14}$ cm^{-2} (**Fig. 2b**). The increase of the bunching behavior near $\tau = 0$ with an increase of the excitation power P_{exc} suggests the presence of a metastable state and a three-level configuration typical to solid-state emitters. A linear fit of the power-dependent reciprocal value of τ_1 extracted from $g^{(2)}(\tau)$ fit reveals the lifetime of the excited state at $P_{exc} = 0$ is about 0.71 ns.

In conclusion, we obtained single-photon emitters in AlN through Zr ion implantation and performed initial characterization of their photophysical properties. A correlation between the implantation fluence and SPE density has also been established. Future work on SPEs in AlN will focus on revealing the defect structure and addressing their spin properties.

References

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