CARS microscopy of diamond needles

Andrej Dementjev[1], Renata Karpicz[1], Bo Xu[2], Sergei Malykhin[2], Yuri Svirko[2], Polina Kuzhir[2]

[1]Department of Molecular Compounds Physics, State research institute Center for Physical Sciences andTechnology, Vilnius, Lithuania
[2]Department of Physics and Mathematics, Center for Photonics Sciences, University of Eastern Finland,Joensuu, Finland
yuri.svirko@uef.fi

Exceptional thermal conductivity and broadband transparency1 has made diamond a material of choice for numerous applications spanning from high-power electronics to the hostile environment sensing. Diamond has recently emerged as a particularly promising platform for sensing, which employs a long spin coherence time of color centers at room temperature [1]. Quantum sensing devices can employ diamond needles (DNs), which can be fabricated by plasma enhanced chemical vapor deposition (CVD) [2]. At the length of several micrometers, the DN enriched with nitrogen–vacancy (NV) and silicon-vacancy (SiV) centers provides the efficient collection of PL photons from its tip having apex curvature of 2-10 nm thus enabling a sensing with nanoscale spatial resolution [3].

It is worth noting that the symmetry and homogeneity of the crystalline field in the vicinity of the color center determines quantum sensing performance. However, the important question on the DN apex crystallinity remains unanswered. Specifically, distortion of the crystal lattice over the DN length may complicate quantum applications, which essentially rely on the prescribed orientation of color centers within unit cell of the diamond lattice. Moreover, sp^2-hybridized carbon atoms in the vicinity of the apex will scatter or absorb photons emitted by a color center thus reducing signal-to-noise ratio.

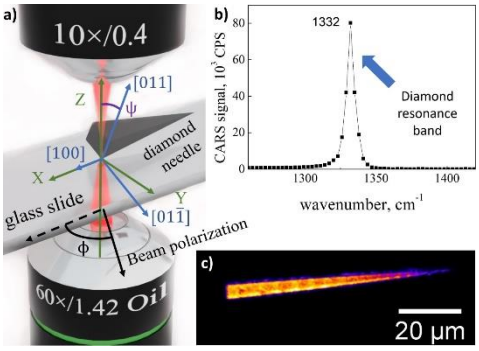

Fig. 1 (a) CARS microscope setup. The orientation of the polarization azimuth of the collinear pump and Stokes beams is determined by angle ϕ, which is controlled by rotating an achromatic λ/2 plate placed before the oil objective. Angle ψ between the slide normal and [011] crystallographic axis of diamond defines orientation of the needle on the glass slide. Cartesian laboratory axes {XYZ} are also shown. In the polarization-resolved experiment, the analyzer was placed after the objective collecting the anti-Stokes signal. (b) The spectrum of the CARS signal in counts per second. (c) CARS image of the tip section (100×30 μm^2) of the needle recorded at $\omega_P - \omega_S$ = 1332 cm^{-1}.

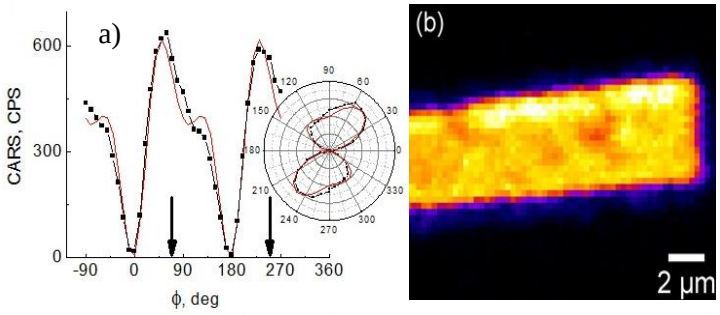

Fig. 2. b) CARS signal as a function of ϕ measured at the analyzer orientation of α =70° (black arrows). The solid red lines show fitting at $|a|^2 = 17400$, ψ = 40° where a is the ratio of the resonant and non-resonant third-order susceptibilities of diamond and ψ defines orientation of the needle on the substrate. b) CARS image of the DN.

The performed measurements [4] showed that a very strong Raman-like band at 1332 cm^{-1} makes it possible to employ CARS microscopy for visualization of the crystallinity over the several micron long diamond needle by exploring the pronounced dependence of the CARS signal on the mutual orientation of the polarization azimuth and diamond crystallographic axes. By mapping CARS signals over the whole needle we demonstrate that the needle is a single crystal, i.e. the quality of the diamond lattice and orientation of the crystallographic axes are the same in both several micron thick base and a nanoscale thin tip of the needle.

The measured polarization-sensitive CARS spectra are well described in terms of the third-order nonlinear optical susceptibility of the diamond. High crystal quality of DNs was verified by showing that FWHM of the SiV color centers emission does not depend on the color center position in the needle. The developed polarization-resolved CARS microscopy technique, which is capable of mapping diamond crystallinity with micrometer precision, can complement the local high-resolution electron microscopy and X-ray diffraction usually applied for the millimeter-scale crystals for diamond diagnostics.

[1] E.D. Herbschleb, et al, Nat. Commun. 10(1), 3766 (2019).
[2] A.N. Obraztsov et al, Diam. Relat. Mater. 18(10), 1289 (2009).
[3] S. Malykhin, et al," Diam. Relat. Mater. 125, (2022).
[4] A. Dementjev et al, Appl. Phys. Lett, 124, 211903 (2024)