Materials Research Bulletin

journal homepage: <www.elsevier.com/locate/matresbu>

# Superemission of Cr nanolayers

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#### A R T I C L E I N F O

## A B S T R A C T

Article history: Received 11 September 2015 Received in revised form 14 December 2015 Accepted 28 December 2015 Available online 3 January 2016

Keywords:

A. inorganic compounds B. metals C. nanostructures D. ptical materials E. thin films

# 1. Introduction

High interest in nanostructured materials and nanolayers arose about 30 years ago, due to unique physical and chemical properties of such materials. Many research reports on synthesis, characterizations and physical and chemical properties of such materials were published  $[1-4]$ . Interestingly, quantum confinement was observed in different nanostructured materials [5–[9\]](#page--1-0). Earlier Khmelinskii and Makarov found [\[10\]](#page--1-0) that metal nanolayers demonstrate interesting optical properties. In particular, Co and Ni nanolayers deposited on fused silica have discrete optical absorption spectra, with the band count and location dependent on the nanolayer thickness. Such nanolayers also produce strong emission when excited by an external light source. These systems should exhibit superemission and operate as lasers, as the population inversion may be created in such multilevel systems upon excitation. In other words, such systems could operate as nanolasers, lasers with nanoscale dimensions.

The nanolaser concept was developed by Mark Stockman at Georgia State University in 2003 [\[11\]](#page--1-0). These tiny lasers can be modulated quickly and, combined with their small footprint, this makes them ideal candidates for on-chip optical computing. The intense optical field of such a laser also enables non-linear optical phenomena and surface-enhanced Raman scattering (SERS) [\[12\],](#page--1-0) and therefore paves the way toward integrated nanophotonic circuitry [\[13\]](#page--1-0). A working room-temperature nanolaser "based on

<http://dx.doi.org/10.1016/j.materresbull.2015.12.047> 0025-5408/Published by Elsevier Ltd.

We continue our studies of metal nanolayers started earlier [1]. Presently, we investigate absorption, emission and superemission in Cr nanolayers. We provide experimental estimates of the energy density and power density of superemission of Cr nanolayers, along with estimates of the diffraction-limited light divergence. We report that the divergence of the superemission along the direction normal to the nanolayer corresponds to that of a point source, with the divergence angle of ca. 0.2 rad. Additionally, we report surprising long-range transfer of the excitation energy in Cr metal films, at macroscopic distances of 1 cm.

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three-dimensional Au bowtie nanoparticles supported by an organic gain material," was reported that should be suitable for inclusion in photonic circuit architectures [\[14\]](#page--1-0). Recently, a thresholdless laser and a room temperature nanolaser were successfully tested, both using plasmonic nanoscale coaxial structures [\[15\]](#page--1-0).

Earlier we investigated optical properties of Co and Ni nanolayers [\[10\]](#page--1-0), finding that such nanolayers demonstrate discrete absorption spectra and strong photoluminescence. We found that the absorption band density grows with the nanolayer thickness. These studies show novel and promising ways to the development of photosensors and nanolasers.

Presently we continue our studies of the metal nanolayers [\[10\],](#page--1-0) addressing optical absorption and emission properties of Cr nanolayers, including their superemission. We found that such metal nanolayers have discrete absorption spectra [\[10\]](#page--1-0). Presently, we report the optical properties of Cr nanolayers of different thickness, deposited on the fused silica substrate. Such nanolayers exhibit strong superemission due to population inversion between the pumped excited state and lower-lying excited states. The superemission yield, pulse energy, energy density and power density were estimated, along with the beam divergence.

# 2. Experimental

Experimental measurements of the absorption, emission and superemission spectra were carried out on model devices using Cr metal nanofilms  $12.5 \times 12.5$  mm<sup>2</sup> in area with variable nanothickness. The Cr nanolayers were deposited on fused silica substrates. Two different optical setups were used, to be presented separately.







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### 2.1. The experimental methods and techniques

Commercial square fused silica substrates  $12.5 \times 12.5 \times 1.0$  mm<sup>3</sup> in size (Valley Design Corp.) were used to deposit Cr layers with the required thickness in the nm range. Commercial Cr targets (Sigma/ Aldrich) were used to produce nanolayers on a commercial sputtering/thermo-evaporation Benchtop Turbo deposition system (Denton Vacuum). The film thickness was controlled by XRD. The XPert MRD system (PANalytic) was calibrated by standard nanofilms of the same material. The estimated absolute uncertainty of the Cr nanolayer thickness measurement was 7%; the relative uncertainties were much smaller, determined by the shutter opening times of the deposition system. Roughness and structure of the sample were controlled by atomic force (5500 Atomic Force Microscope, N9410S Keysight Technology), scanning electronic (JSM-7800F Extreme-resolution Analytical Field Emission SEM, Jeol Inc.) and transmission electronic (JEM-2200FS Transmission Electron Microscope, Jeol Inc.) microscopy.

Absorption spectra were recorded on a Hitachi U-3900H UV– Visible Spectrophotometer. The spectral peak maxima and widths were located using the PeakFit software or Sigmaplot-10. The substrates with the deposited Cr nanolayers were reproducibly installed into a holder made of blackened Al metal.

Fig.1 shows the diagram of the experimental setup used for the measurements of the emission spectra. High-pressure mercury lamp provided the excitation (ESI1200 100W, Planet Bulb Inc.), the emission was collected by the lens system as shown in Fig. 1, using a water-cooled glass filter passing UV radiation of the 313 nm Hg emission band. The filtered UV light was exciting a narrow strip of the nanolayer adjacent to one of the sides of the square, limited by a slit sized  $12.5 \times 1.0 \text{ mm}^2$ . The emission was collected from the



Fig. 1. Block diagram of the experimental setup used to study the sample emission. (A) The photographic setup: 1—the high-pressure mercury lamp (100W); 2—the lens with the focal length  $f_1 = 10$  cm; 3—the lens with  $f_2 = 10$  cm; 4—the lens with  $f_3 = 3$  cm; 5—the planar mirror; 6—the fused silica substrate; 7—the Cr nanolayer; 8—the photo camera; 9—the collimating slit;  $(B)$  The spectroscopic setup: 1–7 are the same as in A; 8—the collimator lens with  $f_4$  = 30 cm; 9—the monocromator; 10 the PMT; 11—the collimating slit.

remaining  $12.5 \times 11.5$  mm<sup>2</sup> of the sample area, separated from the excited zone by a non-transparent shield. The sample emission was either photographed to evaluate its intensity distribution over the sample or focused upon the entrance slit of the monochromator (MonoSpec/50 from Thermo Jarrell Ash). The light exiting the monochromator was detected by a photomultiplier (PMT, H10426 from Hamamatsu Inc.), powered at 1500V by a high-voltage power supply (Model PS350 from Stanford Research Inc.). The output signal of the PMT was recorded by an I/O data acquisition board (PCI-6034E DAQ, National Instruments), programmed in the LABVIEW environment that run on a Dell PC, with the resulting spectra processed on the PC.

[Fig.](#page--1-0) 2 shows the diagram of the setup used to record the superemission of the samples. Excitation was provided by a XeCl excimer laser (LPX-200, Lambda Physics) focused on the sample by a cylindrical lens as shown in [Fig.](#page--1-0) 2. The superemission was collected by a spherical lens with  $f = 30 \text{ cm}$  and after passing through a neutral diffuser and monochromator (MonoSpec/50, Thermo Jarrell Ash) was detected by the photodiode (PD; DET10A Si Biased Detector from THORLABS). The signal from the PD was stored by the data acquisition system based on a PC computer and using a digital oscilloscope (WaveSurfer 400 series, LeCroy), digital delay generators (DG-535, Stanford Research), boxcar integrators (SR-250, Stanford Research), a fast amplifier (SR-240, Stanford Research), and a computer interface board (SR-245, Stanford Research). The emission spectra were scanned at  $1 \text{ cm}^{-1}$  resolution.

### 2.2. The sample preparation procedure

The surface of the fused silica substrates was cleaned by ultrasonication with diamond nanopowder (100 mg) in isopropanol (50 ml) for 30 min at room temperature. Then the substrates were retrieved, washed by 100 ml of isopropanol, dried at 80 $\degree$ C for 10 min, and transported to the sputtering equipment. The Cr nanolayers of different thickness were deposited at the background pressure of about  $10^{-7}$  Torr with the substrate at 300 °C. Six samples were prepared with the Cr nanolayer 7.7, 9.4, 11.8, 12.3, 16.5, and 18.7 nm thick. Since deposition of Cr nanolayers was performed in high vacuum, the formation of any Cr oxides during deposition can be neglected. After deposition, the sputtering chamber was filled with high-purity  $N_2$  gas. Then the chamber was opened and the substrate transported in the hermetic container filled with high-purity  $N_2$  gas. All measurements were carried out in high-purity  $N_2$  atmosphere. Therefore, we assume that no Cr oxides were formed during the experiments. We shall also discuss the HRTEM results, showing that no structures that could be related to the formation of Cr oxides could be observable. The emission and superemission spectra of the 6 film samples were recorded.

#### 2.3. Superemission: time-resolved experiments and spectra

Since all of the superemission experiments were carried out with pulsed excitation (ca. 20 ns pulse duration), a time-resolved data acquisition system was used to collect the superemission signal. The excitation pulse was split at a  $45^{\circ}$  fused quarts plate into two parts, with 93% and 7% of the total pulse energy. The 7% pulse passed through a ND filter and was detected by a fast photodiode (PD1), with the signal used for external trigger of the digital delay generator (DG). The DG generated a TTL pulse with zero delay, used to trigger the digital oscilloscope. Thus, the oscilloscope sweep was synchronized with the pump laser pulse. The 93% pulse was directed to the sample surface, with the sample superemission detected by a fast photodiode PD2 or by a photomultiplier tube (PMT). The signal from the latter was recorded by the digital oscilloscope, with accumulation over at least 512 laser shots.

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