Vacancy clusters at nanoparticle surfaces

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Nanoparticles are important because they exhibit unique properties due to size effects, quantum tunneling, and quantum confinement. These novel properties will be important for new generations of semiconductor and optical communication devices that are being driven to nanoscale dimensions. As particle sizes are reduced to the nanometer scale, defects at the interfaces of nanoparticles can dictate the bulk properties since the surface-to-bulk ratio increases significantly at the nanoscale. In this work, we report our studies of vacancy defects at the surfaces of Au nanoparticles embedded in MgO using a combination of beam positron lifetime spectroscopy, which tells the size of the vacancy cluster, and coincidence measurements of Doppler broadening annihilation radiation, which reveal elemental environment of the vacancy clusters.

Gold nanoparticles are formed in the near surface region of single crystals of MgO using a combination of ion implantation and thermal annealing. Positron lifetimes were measured as a function of positron energy and analyzed using CONTIN and POSFIT. The measured lifetimes are matched with those for various defects in MgO, calculated using the insulator model developed by Puska and Maklinen. Results show that clusters of four atomic vacancies in gold nanocomposites with magnesia matrix. S parameter profiles have been measured for Au/MgO system before and after Au nanoparticle formation. A low-S parameter “valley”, coincident with the Au nanoparticle range, is superimposed on a broad band of high S parameters yielded by vacancy defects generated by implantation and annealing. This suggests some of these vacancy clusters locate at the surfaces of Au nanoparticles.

To confirm vacancy clusters at nanoparticle surfaces, we have conducted two detector coincidence measurements of Doppler broadening annihilation radiation, a system similar to that developed by Asoka-Kumar and Lynn. Results show that high momentum components for positrons annihilating at the Au/MgO nanocomposites display two enhanced bands at 514.3 keV and 518.5 keV, which are very similar to those measured for a pure Au film. These bands are different from that of the MgO layers. The difference is largely interpreted as the Au neighbors of the vacancy clusters, confirming the presence of the clusters on Au nanoparticle surfaces.

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