Journal of Physics: Conference Series **572** (2014) 012024 doi:10.1088/1742-6596/572/1/012024

Carbon encapsulation of magnetic metal nanoparticles: correlation between nanoscale structure of carbon matrix and electromagnetic properties

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Abstract. A comparative analysis of modification of Raman spectra of hydrogenated amorphous carbon films containing encapsulated cobalt nanocrystals, their EM radiation losses and distribution function of nanosized fragments of graphene flatness constituting skeleton of amorphous carbon is performed. For this purpose, a model of confinement of graphene phonons by its fragments is developed, for two types of phonons contributing to the D and G peaks of Raman spectra, as a function of Co content.

1. Introduction

Metal nanoclusters are promising materials for creation on their basis magnetic media for ultra-high density recording and electromagnetic (EM) radiation absorbers. The ensembles of encapsulated clusters show emerging quantum physical effects. Methods of introduction of metal clusters in carbon matrix, also termed as “encapsulation” methods, are of two types. In the first method, metallic clusters are encapsulated into carbon cells. In the second method nanoclusters are embedded into a matrix of amorphous carbon. This technique is less expensive, simpler and more effective than the first one and compatible with thin film technologies for microelectronics.

2. Experimental

Films of amorphous carbon modified with Co (a-C:H(Co)) were produced by simultaneous sputtering of graphite and cobalt targets by Argon (80%) –Hydrogen (20%) plasma in DC magnetron. The films were deposited on Si substrates (for Raman experiments) and aramide tissue (for microwave absorption experiments). Cobalt content was varied by a change of surface of sputteing Co target. Its concentration in the produced films was controlled by Rutherford backscattering method as published elsewhere [7].

Intensity of Raman scattering (figure.1) was measured by SPEX RAMALOG spectrometer using unpolarised light with 488 nm wavelength operating in backscattering geometry.

Microwave absorption experiments were performed using technique published elsewhere [6].

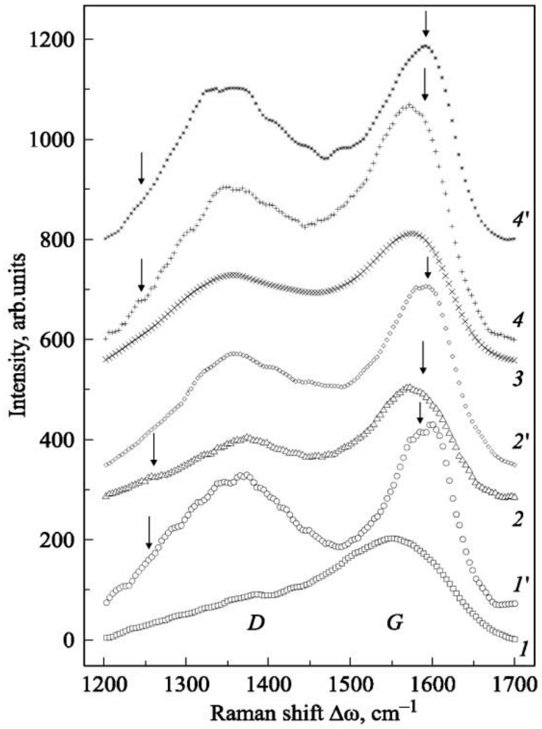

Figure.1. Raman spectra of a-C:H films (1,1’) and a-C:H<Co> (2, 2’,3,4,4’) with different Co content: (1-4) – before thermal annealing and (1’,2’,4’) – after thermal annealing: [Co/C]: 1, 1’—0; 2, 2’:0.35; 3—0.5; 4, 4’—0.73

3. Results and discussion

We used data for microwave absorption published elsewhere [6] (figure.2) for analysis. The dependence of losses for microwaves measured at 10 GHz on Co content is presented in figure.3. It is seen that the dependence attains a maximal value for a certain Co content, for approximately 33 at.%.

These data for microwave absorption were obtained for the samples whose Raman spectra were investigated simultaneously and partially published in paper [7] and presented in figure.1.

Typical Raman spectrum of a-C:H(Co) film is presented in figure.4. The overall shape of the contours was not changed drastically with variation of Co concentration while positions of peaks D and G bands and their ratio were changed.

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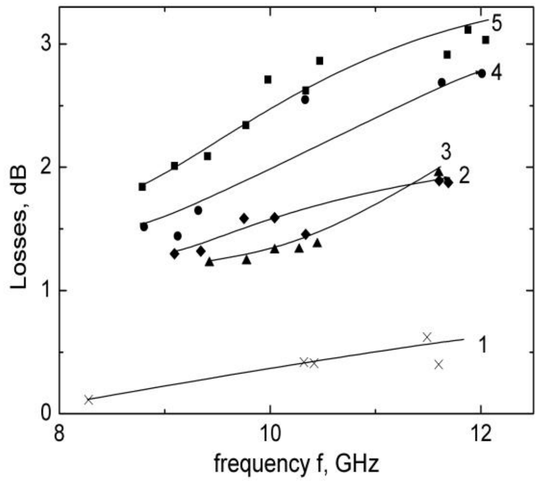

Figure.2. RF losses L of electromagnetic waves in four-layer a-C:H Co covers on aramide tissues. The a-C:H Co film thickness on aramide filaments and the Co content are: 1— four layers of the aramide tissue ; 2— 1.05 mm, 32-at.% Co; 3— 0.95 mm, 39-at.% Co; 4— 0.95 mm, 45-at.% Co; 5— 2.1 mm, 39-at.% Co;

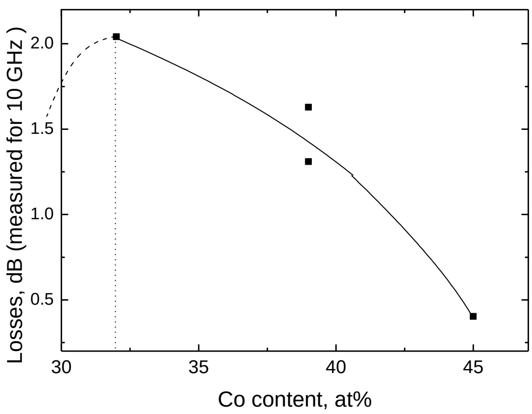

Figure.3. Black squares stand for the dependence of losses of microwaves measured at 10 GHz on Co content for different a-C:H(Co) samples. The curve shows the trend.

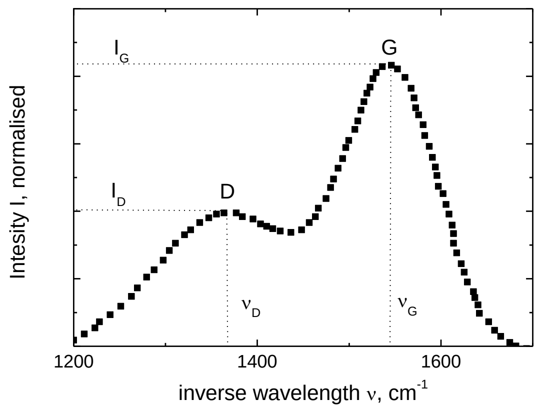

Figure.4. Typical Raman spectrum of a-C:H(Co) spectrum. Intensities of D and G bands and their spectral positions are marked as I_D, I_G and ν_D, ν_G correspondingly.

A quite natural to associate the effect of the intensity changes with variation of the mean size of graphene fragments constituting skeleton of carbon matrix. This variation may occur as a result of presence of Co atoms that may be scattered in the matrix. For the estimation of the mean size we used results presented in paper [8] where a correlation between size of graphene fragment and I_D/I_G ratio was investigated. The result is presented in figure.5.

The experimental dependence of I_D/I_G ratio obtained from figure.2 is shown in figure.6 together with restored diameter of the mean graphene fragments constituting matrix of a-C:H(Co) matrix. The restoration was performed using I_D/I_G ratio and dependence presenting

in figure.4. As is easily seen, the mean size of graphene fragment slightly increases with increase of concentration of Co, while the overall trend attains saturation at higher Co values. The Co/C ratio where the trend attains saturation is about 0.5 (that corresponds to almost 30 at.% of Co). This concentration corresponds to a stechiometric formula CoC_2 for the considering substance. At the same time, as is seen in figure.6, the mean diameter of graphene fragment equals to approximately 11 Å.
This value correlates with concentration of Co where losses of microwaves are maximal, as figure.3 shows. As one might expect a molecular cluster with approximate CoC_2 formula might be responsible for absorption of microwaves and might be associated with Co-intercalated fragments of graphene either single or layered fragments of graphene.
We have linear dependence between the number of the rings and the approximate diameter of fragment (figure.7), so the mean number of benzene rings, M, that cover the compact graphene cluster with mean size of 11 Å, might be restored using that dependence.
It is seen that M=20 for the case.

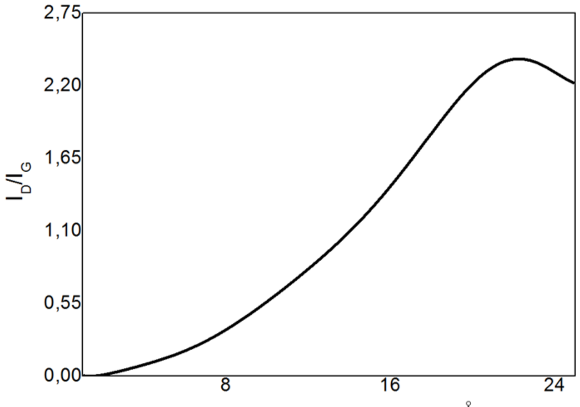

Figure.5. The dependence of I_D/ I_G ratio on the size of graphene fragment.

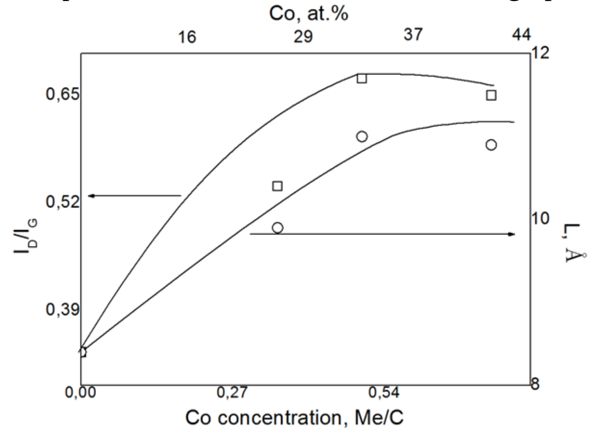

Figure.6 Open squares portrait experimental values for ration of intensities of D and G bands for a-C:H(Co) samples v.s. concentration of cobalt. Open circles show calculated values of the mean diameter of graphene fragments calculated using open square data and the function presenting in Figure 5. Full lines demonstrate corresponding trends.

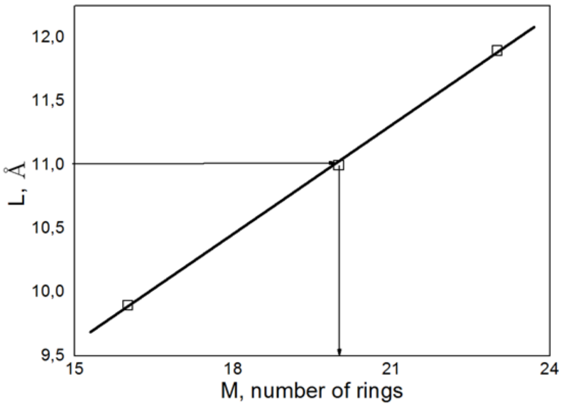

Figure.7 Dependence of mean size of graphene fragment on the number of six-member rings constituting it (results are collected from [7]).

A model of graphene fragment intercalated with Co is shown in figure. 9

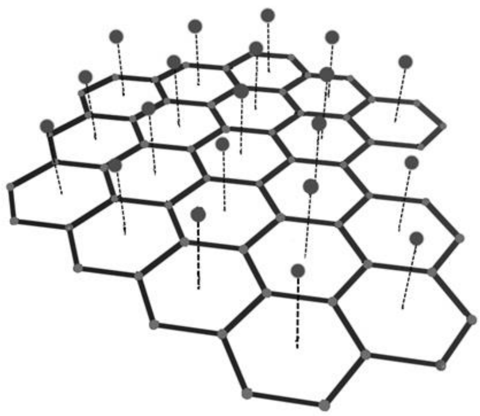

Figure. 9 A model of graphene fragment intercalated with Co.

4. Conclusions

Correlation in between microwave absorption and Raman spectra of a-C:H(Co) was shown. Maximum absorbtion is reached for the concentration of Cobalt approximately 30% that corresponds to the mean diameter of graphene fragment approximately 11 Å.
Fragments of Co-intercalated graphene might be responsible for this effect along with massive cobalt nanoclusters.

5. References

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