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Revision of optical properties of solids based on the reverse Monte Carlo analysis of reflection electron energy loss spectroscopy spectra

K. Tőkési,[1,2,*] L.H. Yang,[3] D. Bo,[4] and Z.J. Ding[3]
[1] *Institute for Nuclear Research, Hungarian Academy of Sciences (ATOMKI), Debrecen, Hungary*
[2] *ELI-ALPS, ELI-HU Non-profit Ltd., Dugonics tér 13, H-6720 Szeged, Hungary*
[3] *Hefei National Laboratory for Physical Sciences at Microscale and Department of Physics, University of Science and Technology of China, Hefei 230026, Anhui, P.R. China*
[4] *Research and Services Division of Materials Data and Integrated System, National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, Japan*

*corresponding author's e-mail: tokesi@atomki.mta.hu

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We present high precision determination of electron energy loss functions and thereby the optical constants, *n* and *k*, of solids from the measured, high energy resolution reflection electron energy-loss spectroscopy (REELS) spectra, covering the spectral energy range from visible to vacuum ultraviolet. The calculations are based on our recently developed reverse Monte Carlo (RMC) method. The RMC method combines a Monte Carlo modelling of electron transportation for REELS spectrum simulation, including both the elastic and inelastic collisions, with a Markov chain Monte Carlo calculation of parameterized energy loss function, Im [-1/ε(ω)]. We found that our calculated optical data of elements fulfill the sum rules with very high accuracy; therefore, the use of this calculated optical data in material science and surface analysis is highly recommended for further applications.

There is a continuous interest and effort in the determination of optical constants, i.e. the refractive index *n* and extinction coefficient *k*, of solids due to their importance in both fundamental researches and applications. Usually they are determined by optical measurements, and the measured optical data at the long wavelength limit ($q \to 0$) from different sources were compiled by Palik [1] for many elements and compounds. We note that with the demands of scientific development the optic techniques have reached their limitations. The optical measurements are usually performed under atmospheric condition and therefore the surface cleanliness definitely can not guaranteed. Moreover, the reflection experiments are sensitive to the surface roughness. In addition, the energy region of the measured optical constants is directly constrained by the light source and therefore the range is usually very short.

These drawbacks can be overcome by using electron probe techniques, and they can provide an alternative way for deriving information of dielectric response of solid, which is, in principal rather different technique compared with optical methods. In this years a well-established technique based on the reflection electron energy loss spectroscopy (REELS) has been developed [2-4] to obtain optical constants in a rather wide range of energy loss of electrons (i.e., photon energy). The typical energy loss range is 1–100 eV, and the measurements can be performed once or maybe several times under different experimental conditions but with the same spectrometer. Such an ability to derive optical constants in a wide photon energy range with only one spectrum is the main advantage of REELS compared with the optical measurements. In addition, it also holds the opportunity to get the optical constants for nonzero momentum transfers. In deriving

the energy loss function (ELF), Im[$-1/\varepsilon(\omega)$], and thereby the optical constants (n,k), where $\varepsilon = n + ik$ is the complex dielectric function of the solid, from the measured REELS spectra, precise and accurate knowledge of the electron energy loss processes has crucial importance.

Recently a reverse Monte Carlo (RMC) method for the data analysis of REELS spectra has been developed by Da et al. [5] to obtain optical constants in a rather wide range (0-200 eV) of energy loss of electrons with very high accuracy. This RMC method combines the Monte Carlo simulation of REELS spectrum with a Markov chain Monte Carlo sampling of oscillator strength parameters. Electron interaction with a sample is comprised of the electron elastic scattering and the bulk- as well as surface-electronic excitation for electron inelastic scattering. Our Monte Carlo simulation model is based on Mott's cross-sections for electron elastic scattering as calculated by partial wave method and dielectric functional approach for electron inelastic scatterings. During the simulation we used the depth dependent differential inverse inelastic mean free path.

In this work, applying our RMC method, we present high precision determination of electron energy loss functions and thereby the optical constants, n and k, of metals and semiconductors from the measured, high energy resolution REELS spectra [6-8]. As an example, Fig. 1 shows the calculated refractive index n and extinction coefficient k of Fe in the photon energy range of 0-200 eV, in comparison with Palik's database of optical constants. While we found perfect agreement with the previous experimental data in the high energy-loss region, our data join smoothly the refractive index n and extinction coefficient k of Palik's database in the absent range of 26–40 eV. The high accuracy of the obtained results are justified with various sum rules, like f - and ps-sum rules. We found that our calculated optical data of elements fulfill the sum rules with very high accuracy; therefore, the use of this calculated optical data in material science and surface analysis is highly recommended for further applications.

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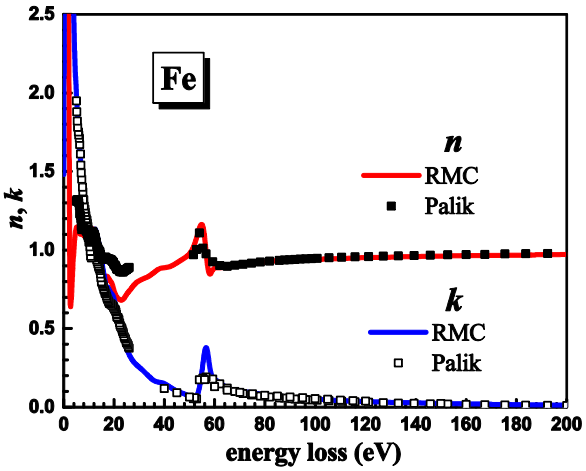

Fig. 1 Comparison on the refractive index n and extinction coefficient k of Fe between the RMC method [6] and Palik's data [1].

References

[1] E.D. Palik, Handbook of Optical Constants of Solids, Academic Press, New York, (1985).

[2] F. Yubero, S. Tougaard, Model for quantitative analysis of reflection-electron-energy-loss spectra, Phys. Rev. B 46 2486 (1992).

[3] T. Nagatomi, T. Kawano, and R. Shimizu, J.Appl. Phys. **83**, 8016 (1998).

[4] W. S. M. Werner, Appl. Phys. Lett. **89**, 213106 (2006).

[5] B. Da, Y. Sun, S.F. Mao, Z.M. Zhang, H. Jin, H. Yoshikawa, S. Tanuma, and Z.J. Ding, J. Appl. Phys. 113 214303 (2013).

[6] H. Xu, B. Da, J. Tóth, K. Tőkési and Z.J. Ding, Phys. Rev. B 95 (2017)195417.

[7] L.H. Yang, M. Menyhárd, A. Sulyok, K. Tőkési, and Z.J. Ding, Applied Surface Science, 456: 99 (2018)

[8] L.H. Yang, M. Menyhard, A. Sulyok, K. Tőkési, Z.J.Ding, Eur. Phys. J. D (2019) 73: 21.