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Infrared Reflectance Kramers–Kronig Analysis by Anchor-Window Technique

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An algorithm for the Kramers–Kronig analysis of the reflectivity spectra, based on an anchor-window technique is presented. The high-frequency asymptote, required for the Kramers–Kronig analysis, is determined by minimizing differences between the Kramers–Kronig-deduced optical constants of a system under investigation and known optical constants measured in a small anchor-window. The algorithm is illustrated by applying it for a reconstruction of the optical conductivity $\sigma(\omega)$ of the fci-ZnMgRE quasicrystals in the spectral range of 0.01–6.5 eV from the experimental IR Fourier-transform reflectivity data and the experimental spectral ellipsometry VIS-UV data. The reliability of the suggested Kramers–Kronig analysis technique is confirmed by independent infrared spectral ellipsometry $\sigma(\omega)$ measurements for fci-ZnMgRE.

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1. Introduction

The infrared reflectance spectroscopy is a main tool for optical investigations in the IR spectral range. The optical parameters of a system under investigation, the dielectric function $\varepsilon(\omega)$ or the optical conductivity $\sigma(\omega)$ (which is directly related with the dielectric function as $\varepsilon(\omega) = 1 + \mathrm{i}4\pi\sigma(\omega)/\omega$) are determined from the reflectivity spectrum $R(\omega)$ making use of the Kramers–Kronig (KK) relations. The KK analysis, however, requires for extrapolations of the experimental data to the low- and high-frequency limits, which usually essentially reduces an accuracy of the analysis.

We present an algorithm of the Kramers–Kronig analysis based on an anchor-window technique. The high-frequency asymptote of the reflectivity spectrum is determined by a minimization of differences between the KK-deduced optical conductivity $\sigma_{\mathrm{KK}}(\omega)$ and the known $\sigma(\omega)$ values, measured a priori in a small anchor-window. The algorithm is illustrated by reconstructing the wide-range, 0.01–6.5 eV, optical conductivity spectrum of fci-ZnMgRE (RE = Y, Ho) quasicrystals.

2. Experimental

The single-grain face-centred icosahedral (fci) $Zn_{62}Mg_{29}Y_9$ and $Zn_{65}Mg_{25}Ho_{10}$ quasicrystals were grown by the liquid-encapsulated top-seeded solution-growth method [1]. Prior to each optical measurement, the optical surfaces of samples were mechanically grind and fine-polished with a diamond paste (10 μm, 5 μm, 1 μm, and 0.25 μm) and an alumina suspension (0.02 μm).

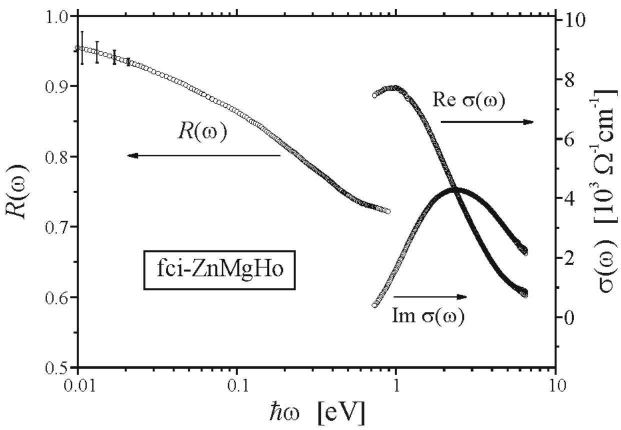

Fig. 1. Experimental fci-ZnMgHo reflectivity $R(\omega)$ and the optical conductivity $\sigma(\omega)$ spectra.

The IR reflectivity $R(\omega)$ spectra (Fig. 1) in the spectral range 0.01–0.9 eV were measured by Fourier transform spectrometer Nicolet 8700. The VIS-UV optical conductivity $\sigma(\omega)$ spectra (Fig. 1) in the spectral range 0.73–6.5 eV were measured by spectroscopic ellipsometry (SE) technique by the rotating analyzer ellipsometer VASE (J.A. Woolam Co, Inc.). Appending the IR reflectivity data by the $R(\omega)$, calculated from the SE VIS-UV data, we obtained the reflectivity spectra in the spectral range 0.01–6.5 eV (see Fig. 2).

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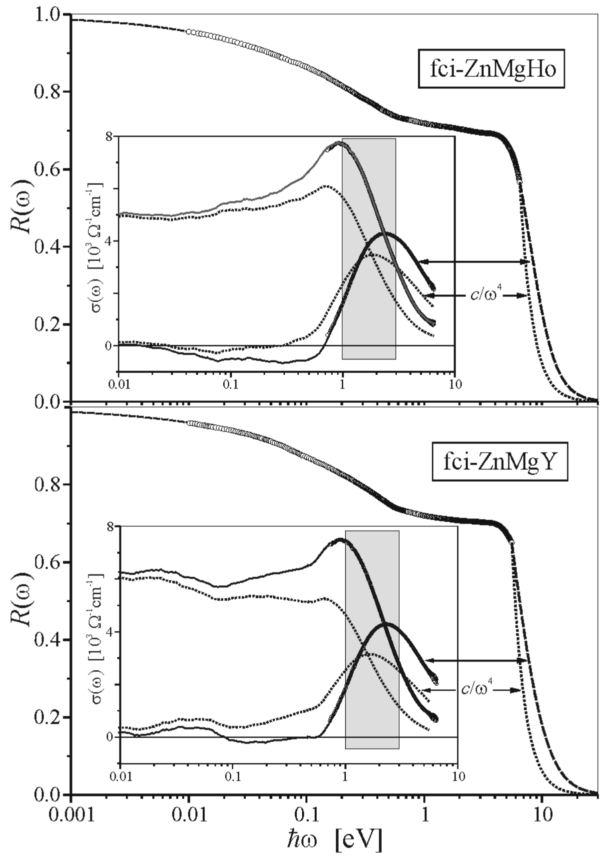

Fig. 2. The experimental reflectivity $R(\omega)$ spectra (dots) and extrapolations by the standard high--frequency asymptote $R_{\rm hf}(\omega) = c/\omega^4$ (dotted curves) and by the anchor-window determined asymptote (dashed curves). Insets present the experimental optical conductivity $\sigma(\omega)$ spectra (dots) and the KK-deduced $\sigma_{\rm KK}(\omega)$ spectra (dotted and dashed curves). Shaded areas in insets indicate the anchor-window.

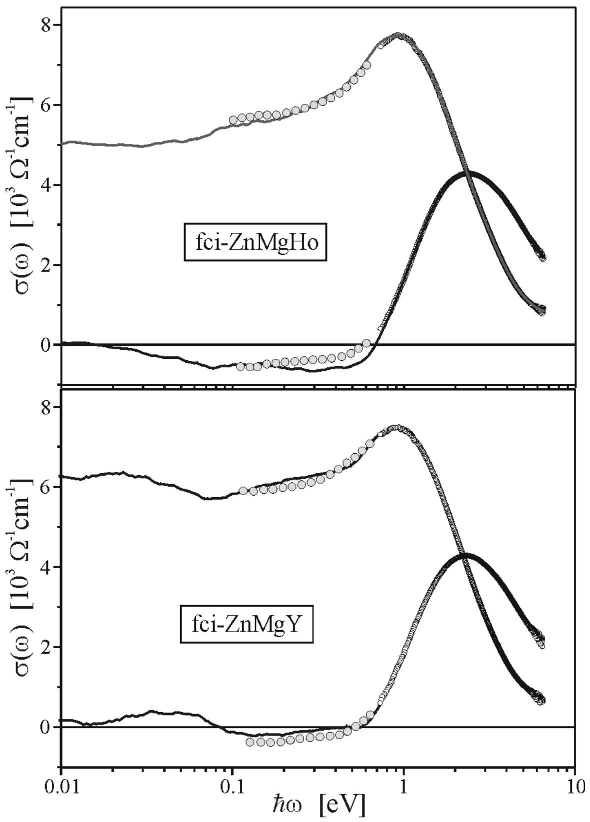

Fig. 3. Experimental optical conductivity $\sigma(\omega)$ spectra (dots) and the $\sigma(\omega)$ spectra deduced by the Kramers–Kronig analysis (curves).

3. Algorithm of the anchor-window technique

In metallic compounds, the low- and high-frequency reflectivity asymptotes, required for the Kramers–Kronig analysis, are usually approximated by the relations

$$R_{\rm lf}(\omega) = 1 - c\sqrt{\omega}\,, \quad R_{\rm hf}(\omega) = \frac{c}{\omega^4}\,. \tag{1}$$

Here the low-frequency asymptote $R_{\rm lf}(\omega)$ corresponds to the Hagen–Rubens law and the high-frequency asymptote $R_{\rm hf}(\omega)$ corresponds to a generic frequency dependence, which settles down at photon energies exceeding characteristic energies of a system under investigation. Making use the $R(\omega)$ extrapolations (1) and of the Kramers–Kronig relation

$$\theta(\omega) = -\frac{\omega}{\pi} P \int_0^\infty {\rm d}\omega' \frac{\ln R(\omega')}{\omega'^2 - \omega^2}\,, \tag{2}$$

one can calculate the phase factor $\theta(\omega)$ of the complex reflection amplitude $r(\omega) = |r(\omega)| \exp({\rm i}\theta(\omega))$. Then, making use of the relation $r(\omega) = [\varepsilon^{1/2}(\omega) - 1]/[\varepsilon^{1/2}(\omega) + 1]$, one can determine the dielectric function and the optical conductivity. Results of the calculations are presented in insets to Fig. 2 by dotted curves. As seen, the optical conductivity $\sigma_{\rm KK}(\omega)$, deduced by the Kramers–Kronig analysis, essentially differs from experimental $\sigma(\omega)$ spectra, presented by dots in Fig. 2 insets.

There are various techniques to improve an accuracy of the KK analysis [2]. We suggest an anchor-window technique, which is a modification of the anchor-point method [3]. The high-frequency asymptote is modelled by the inverse polynomial

$$R_{\rm hf}(\omega) = \left[b_0 + b_1\omega + b_2\omega^2 + b_3\omega^3 + b_4\omega^4\right]^{-1}, \tag{3}$$

the coefficients b_i of which are determined by minimizing differences between the KK-deduced optical conductivity and the experimental $\sigma(\omega)$ values, known in a narrow spectral region — the anchor-window. The error function was chosen in the form

$$\chi = \frac{1}{N} \sum_{n=1}^{N} \left| \frac{\sigma_{\rm KK}[n] - \sigma_{\rm exper}[n]}{\sigma_{\rm exper}[n]} \right|^2, \tag{4}$$

where $\sigma_{\rm exper}[n]$ is the experimental optical conductivity value at the spectral point n, which belongs to the anchor-window, $\sigma_{\rm KK}[n]$ is the KK-deduced value, and N is the number of points within the anchor-window. The low-frequency asymptote of the reflectivity spectrum was approximated by the relation

$$R_{\mathrm{lf}}(\omega) = 1 - a_1\sqrt{\omega} - a_2\omega\,. \quad (5)$$

The a_1 and a_2 coefficients were determined by the least--squares technique, minimizing the error function

$$\chi = \frac{1}{N}\sum_{n=1}^{N}\left|R_{\mathrm{lf}}[n] - R_{\mathrm{exper}}[n]\right|^2,$$

where $N = 5$–10 is the number of the first several spectral points of an experimental reflectivity spectrum.

The high-frequency asymptote $R_{\mathrm{hf}}(\omega)$ and the optical conductivity spectrum $\sigma_{\mathrm{KK}}(\omega)$, deduced by the anchor--window technique, are presented by dashed curves in Fig. 2. As seen, the anchor-window technique essentially improves an accuracy of the Kramers–Kronig analysis.

To check a reliability of the anchor-window technique, we measured the IR optical conductivity of fci--ZnMgRE quasicrystals by the spectroscopic ellipsometry technique. The measurements were carried out by a rotating compensator Fourier-transform ellipsometer IRSE (J.A. Woolam Co, Inc.). Results are presented by enlarged dots in Fig. 3. As seen, the KK-deduced optical conductivity values in the IR spectral range are close to the actual ones. This justifies a reliability of the suggested anchor-window technique.

Acknowledgments

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