# Indication of van Hove singularities in the density of states of ZnMg(Y,Ho) quasicrystals

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We present a low-temperature synchrotron-radiation photoelectron microspectroscopy study of the valence bands of single-grain icosahedral ZnMgY and ZnMgHo quasicrystals. Their valence band photoemission spectra reveal in both cases a simple metal-type behavior with a clear metallic Fermi edge and a sharp pseudogap feature: the spectral intensity exhibits a maximum at $\approx 0.7$ eV below the chemical potential and a subsequent decrease as the Fermi level is approached. The analysis of the pseudogap feature, based on the band-structure hypothesis, shows that it corresponds to the van Hove singularities in the density of states (DOS), which originate from the intersections of the isoenergetic surface with the 222100 and 311111 Bragg planes. The reconstructed density of states agrees in general trends with the theoretical first-principles DOS of hexagonal ZnMgY approximants, and correctly reproduces the experimental value of *i*-ZnMgY electronic specific-heat coefficient.

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## I. INTRODUCTION

The valence-band photoemission (PE) spectra of quasicrystals (QC's) are usually dominated by *d* electrons, e.g., the valence-band structure of the most widely studied quasicrystals *i*-AlCuFe and *i*-AlPdMn is determined by Cu $3d$, Fe $3d$ Pd $4d$, and Mn $3d$ derived states (see, e.g., Ref. 1). In the present paper we present a photoelectron microspectroscopy study of *in situ* cleaved, single-grain, icosahedral ZnMgY and ZnMgHo quasicrystals, which reveals their simple metal-type valence bands at least down to $\approx 5$ eV below the Fermi level $\varepsilon_F$.

The most interesting feature observed in PE spectra of quasicrystals is the presence of a pseudogap. The pseudogap has been predicted theoretically in the linear muffin-tin orbital calculations of the QC energy spectrum,[2–4] as well as on the basis of the ''band-structure hypothesis,''[5,6] which treats the electron subsystem in QC's as a nearly free-electron (NFE) gas, affected by a weak quasiperiodic potential. The pseudogap is assumed to be a generic feature of quasicrystals and was observed in *i*-AlCuFe, *i*-AlPdMn, *d*-AlNiCo, and other quasicrystals (see Refs. 1,7,8, and references therein). The pseudogap in experimental PE valence-band spectra manifests itself as a dip in the density of states (DOS) at the Fermi level, and is usually modeled by the Lorentzian shape as has been suggested by Mori *et al.*[7]

The PE valence-band spectra of *i*-ZnMg(Y,Ho), considered in the present paper, indicate a distinct pseudogap structure in the vicinity of the Fermi level. Compared to measurements on other QC's, the pseudogap in *i*-ZnMg(Y,Ho) quasicrystals is definitely much sharper. We present here an analysis of its structure, which reveals that the observed DOS feature reflects the van Hove singularities predicted in the framework of the band-structure hypothesis.[5]

## II. EXPERIMENT

The icosahedral $Zn_{62}Mg_{29}Y_9$ and $Zn_{65}Mg_{25}Ho_{10}$ single-grain quasicrystals were grown by the liquid-encapsulated top-seeded solution-growth method.[9] The structural perfection of the quasicrystals was confirmed by the sharp, resolution limited, Bragg peaks of their diffraction patterns.

Photoemission measurements were performed with the angle-resolved scanning photoelectron microscope, at beamline BL31 of the Swedish synchrotron-radiation facility MAX-lab (Lund). The specimens investigated were prepared in a form of small, $\approx 0.5$ cm long, rods with $\approx 1\ \mathrm{mm}^2$ cross section. The samples were cleaved *in situ* at $\approx 2 \times 10^{-10}$ mbars and low, $\approx 90$ K, temperature. The narrow width (1.5 $\mu$m) of the incident photon beam of the scanning photoelectron microscope allowed us to focus the beam at normal incidence on the freshly cleaved sample surface for PE measurements carried out with the VG CLAM2 analyser mounted at 47.5°. The cleanliness of the cleaved QC surfaces was routinely monitored following photoemission from the O $2p$ level.[10]

## III. RESULTS AND DISCUSSION

The experimental valence-band PE spectra of the ZnMgY and ZnMgHo quasicrystals, recorded at 44 eV incident photon energy and about 90 K, are presented in Figs. 1(a) and 1(b). As can be seen, the valence-band (VB) bottoms of the quasicrystals are dominated by the Zn $3d$ shallow core level. The PE spectrum of *i*-ZnMgY [Fig. 1(a)] above the Zn $3d$ line shows a featureless, simple metal-type, valence band. The small peak at ca. $-6.5$ eV is a ''ghost'' manifestation of the Mg $2p$ core level (with binding energy of $\approx 50$ eV) due to second-order diffraction from the monochromator grating. The PE spectrum of *i*-ZnMgHo [Fig. 1(b)] shows a distinct complex peak at ca. 6 eV below Fermi level, which can be

 

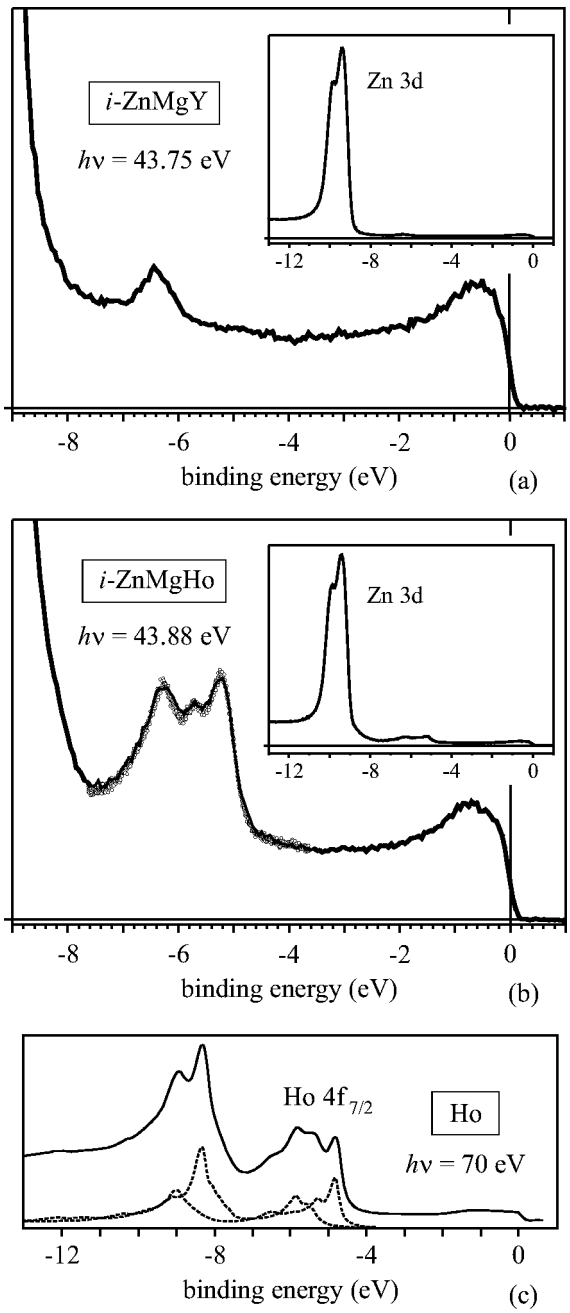


FIG. 1. Valence-band PE spectra of (a) *i*-ZnMgY ($T=93.5$ K) and (b) *i*-ZnMgHo ($T=86.4$ K) quasicrystals, and (c) PE Ho $4f$ line Ref. 11 in pure holmium.

easily identified as the Ho $4f_{7/2}$ complex line from its comparison with the PE spectra of pure holmium,[11] presented in Fig. 1(c).

The observed valence-band PE spectra do not manifest the DOS spikiness, which has been predicted theoretically (e.g., Ref. 2) and which is assumed to be a unique feature of QC's, distinguishing them from other materials. However, one should note that any observation of the spikiness was beyond our reach due to the experimental energy resolution, which for the valence-band PE measurements was typically 0.1–0.25 eV.

Both ZnMgY and ZnMgHo quasicrystal VB spectra show

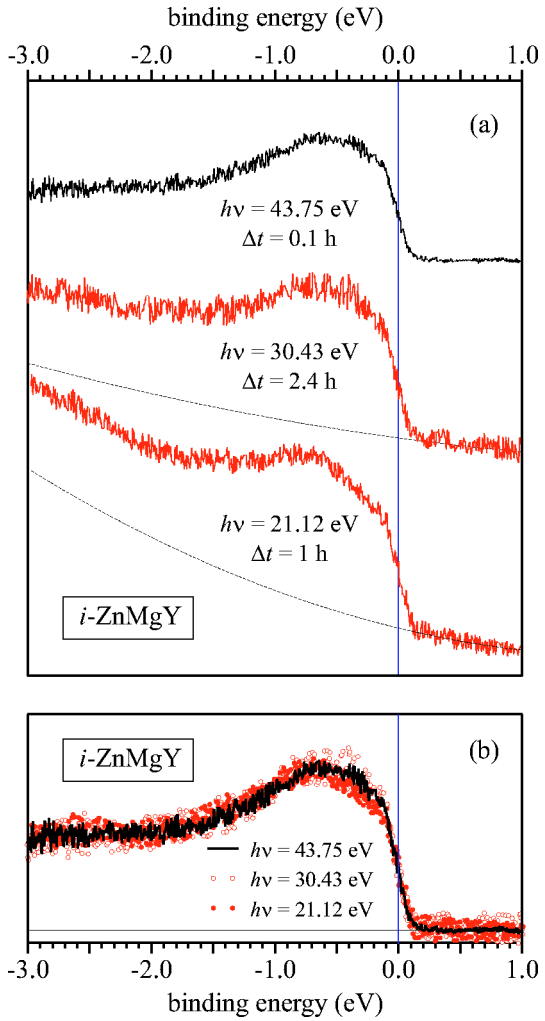


FIG. 2. Experimental *i*-ZnMgY Fermi-level vicinity PE spectra ($T=93.5$ K) recorded at various photon energies (a) without and (b) with background subtraction. (Dashed curves indicate the background. The nominal energy resolution is ca. 0.1 eV. The $\Delta t$ indicates the time interval after the sample cleavage.)

clear metallic Fermi edges and pseudogap features. The spectral intensity exhibits a maximum at $\approx 0.7$ eV below the Fermi level and a subsequent decrease as $\varepsilon_F$ is approached. The experimental Fermi-level vicinity PE spectra, recorded at various photon energies and normalized to the incident photon beam intensity, are presented in Figs. 2(a) and 3. As seen, the pseudogap feature observed remained essentially the same when recorded at various photon energies within the $h\nu$ interval 20–60 eV. If the *i*-ZnMgY pseudogap structure would be due to Y $4d$ derived states, the observed PE spectral feature should strongly increase at low photon energies. Indeed, due to a pronounced Cooper minimum, the Y $4d$ photoionization cross section at $h\nu=21$ eV is ca. 20 times higher than that at $h\nu=44$ eV.[12] However, the feature was not sensitive to the chosen photon energy. We illustrate this separately in Fig. 2(b), which presents the 21-, 30-, and 44-eV PE spectra with subtracted background[18] and normalized to the spectral intensity of the PE shoulder below the pseudogap feature, at $\varepsilon_b < -2$ eV. The apparent stability of

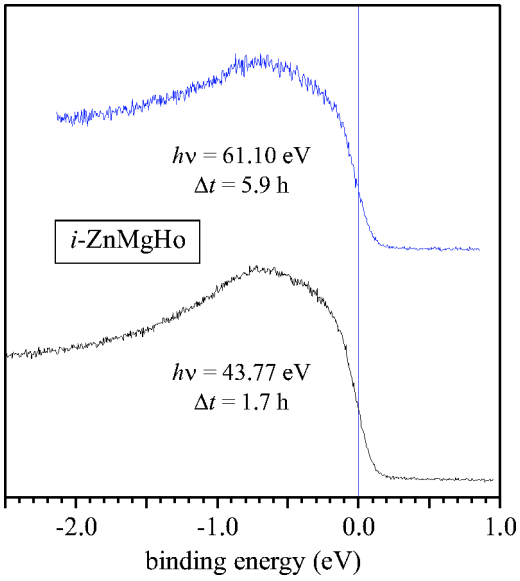


FIG. 3. Experimental *i*-ZnMgHo Fermi-level vicinity PE spectra ($T=93.5$ K) recorded at the photon energies $h\nu=44$ and 61 eV. (The nominal energy resolution is ca. 0.2 eV. The $\Delta t$ indicates the time interval after the sample cleavage.)

the *i*-ZnMgY pseudogap feature and the fact that it was essentially the same in both *i*-ZnMgY and *i*-ZnMgHo quasicrystals, allows us to assume that it is due first of all to *s* (or *sp*) electrons.

To analyze the structure of the near $\varepsilon_F$ spectra we have performed the convolution procedure

$$I(\varepsilon_b)=\int_{-\infty}^{+\infty} dx R(x) f(x) G(x-\varepsilon_b), \tag{1}$$

where $I(\varepsilon_b)$ is the observed spectral intensity, the $R(x)$ function is proportional to the density of states, $f(x)$ is the Fermi-Dirac distribution function, and $G(x-\varepsilon_b)$ is the experimental resolution Gaussian function,

$$G(x-\varepsilon_b)=\frac{1}{\sqrt{2\pi}s}\exp\left(-\frac{(x-\varepsilon_b)^2}{2s^2}\right). \tag{2}$$

The standard deviation of the Gaussian curve, *s*, at around 90 K, is essentially determined by the overall energy resolution of the photoelectron microscope. Analyzing the PE Fermi edge spectra of the reference gold sample at this temperature, we have determined *s* to be 0.1 eV (at $h\nu=44$ eV);[10] this corresponds to a full width at half-maximum of the Gaussian curve of $\sqrt{8\ln 2}s=0.23$ eV.

Assuming that the valence bands of ZnMgY and ZnMgHo quasicrystals are determined by *sp* electrons, one can model their density of states in the framework of the NFE approximation, which predicts van Hove singularities due to intersections of the isoenergetic surface with Bragg planes. When analyzing the pseudogap structure, it is convenient to present the density of states in the form

$$R(\varepsilon)=R_0(\varepsilon)[1+P(\varepsilon)], \tag{3}$$

where $R_0(\varepsilon)$ is the "normal" DOS, without the pseudogap structure, and $P(\varepsilon)$ is the deviation from the normal DOS. Within the NFE approximation the $P(\varepsilon)$ function can be calculated analytically,[13]

$$P(\varepsilon)=P_{\boldsymbol{g}}(\varepsilon)=N\left(\frac{x_{\max}-x_{\min}}{2\sqrt{\varepsilon/\varepsilon_0}}-1\right). \tag{4}$$

Here $N$ is the multiplicity of Bragg planes, the number of corresponding equivalent Brillouin zone faces in usual crystals, $\varepsilon_0=\hbar^2(\frac{1}{2}g)^2/2m$ is the free-electron energy at the $\boldsymbol{k}=\frac{1}{2}\boldsymbol{g}$ wave vector, $\boldsymbol{g}$ is the reciprocal lattice vector, which corresponds to the Bragg planes, $x_{\min}$ and $x_{\max}$ are defined by the following expressions:

$$x_{\min}=1-[1+\varepsilon/\varepsilon_0+\sqrt{4\varepsilon/\varepsilon_0+(\Delta/2\varepsilon_0)^2}]^{1/2}, \tag{5}$$

$$x_{\max}=\begin{cases}1-[1+\varepsilon/\varepsilon_0-\sqrt{4\varepsilon/\varepsilon_0+(\Delta/2\varepsilon_0)^2}]^{1/2}\\ 1\\ 1+[1+\varepsilon/\varepsilon_0-\sqrt{4\varepsilon/\varepsilon_0+(\Delta/2\varepsilon_0)^2}]^{1/2}\end{cases}, \tag{6}$$

for $\varepsilon<\varepsilon_0-\frac{1}{2}\Delta$, $\varepsilon_0-\frac{1}{2}\Delta<\varepsilon<\varepsilon_0+\frac{1}{2}\Delta$, and $\varepsilon>\varepsilon_0+\frac{1}{2}\Delta$, respectively, where $\Delta=2|V_{\boldsymbol{g}}|$ is the pseudogap width and $V_{\boldsymbol{g}}$ is the pseudopotential.

Although the reciprocal quasicrystal lattice vectors $\boldsymbol{g}$ fill the reciprocal space densely, not all of them are of physical importance. The important ones can be determined from the QC diffraction patterns. The Fermi wave vectors $k_F=1.51\times10^8$ cm$^{-1}$ and $k_F=1.49\times10^8$ cm$^{-1}$ of *i*-ZnMgY and *i*-ZnMgHo, respectively,[19] lie in the close proximity of the 222100 and 311111 Bragg planes, whose corresponding reciprocal lattice vectors are $\frac{1}{2}\boldsymbol{g}_{222100}=1.53\times10^8$ cm$^{-1}$ and $\frac{1}{2}\boldsymbol{g}_{311111}=1.59\times10^8$ cm$^{-1}$ (in both quasicrystals), as deduced from experimental Debye-Scherrer diffractograms. There are 60 equivalent 222100 Bragg planes and twelve 311111 ones, which make up the effective Brillouin zone, alias the Jones zone, depicted in Fig. 5(a). The *X* and *Z* points, indicated on the Jones zone faces, correspond to the $\frac{1}{2}\boldsymbol{g}_{222100}$ and $\frac{1}{2}\boldsymbol{g}_{311111}$ vectors.

The electron isoenergetic surface is affected by its intersections with the Jones zone faces. Assuming that the intersections do not overlap, one can express the deviation from the normal DOS as

$$P(\varepsilon)=\int_{-\infty}^{+\infty} d\varepsilon'[P_{222100}(\varepsilon')+P_{311111}(\varepsilon')]\times\frac{1}{\sqrt{2\pi}\Gamma}\exp\left(-\frac{(\varepsilon'-\varepsilon)^2}{2\Gamma^2}\right), \tag{7}$$

where the $P_{222100}(\varepsilon)$ and $P_{311111}(\varepsilon)$ functions correspond to the partial contributions of the 222100 and 311111 faces and are defined by expression (4). The Gaussian convolution employed accounts for a possible intrinsic broadening of the QC energy features. Formulas (4)–(7) determine the density of states at given values of the pseudogap widths $\Delta_{222100}$ and $\Delta_{311111}$, and of the broadening parameter $\Gamma$. The calculations of the PE spectral intensity (1) were performed assum-

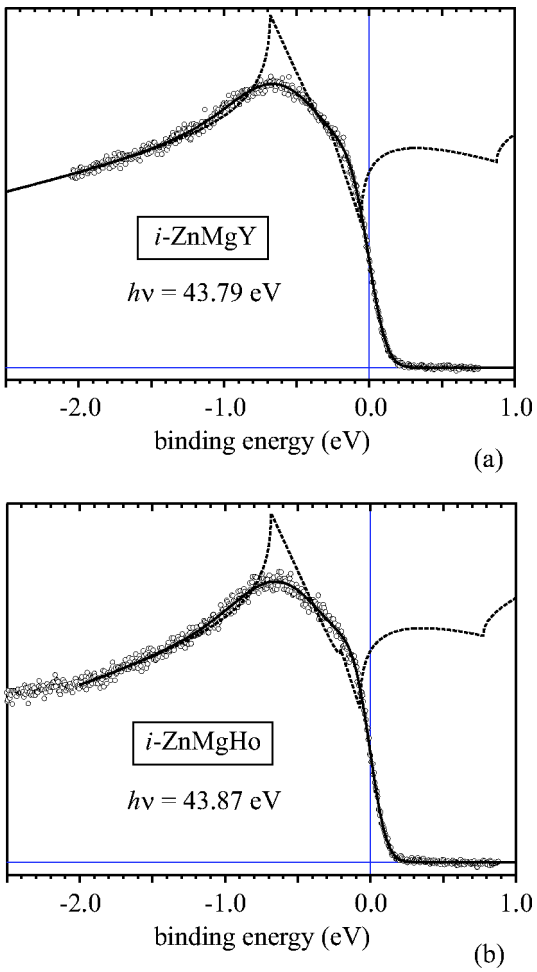


FIG. 4. Near Fermi edge PE spectra of *i*-ZnMgY ($T=93.5$ K) and *i*-ZnMgHo ($T=86.4$ K).

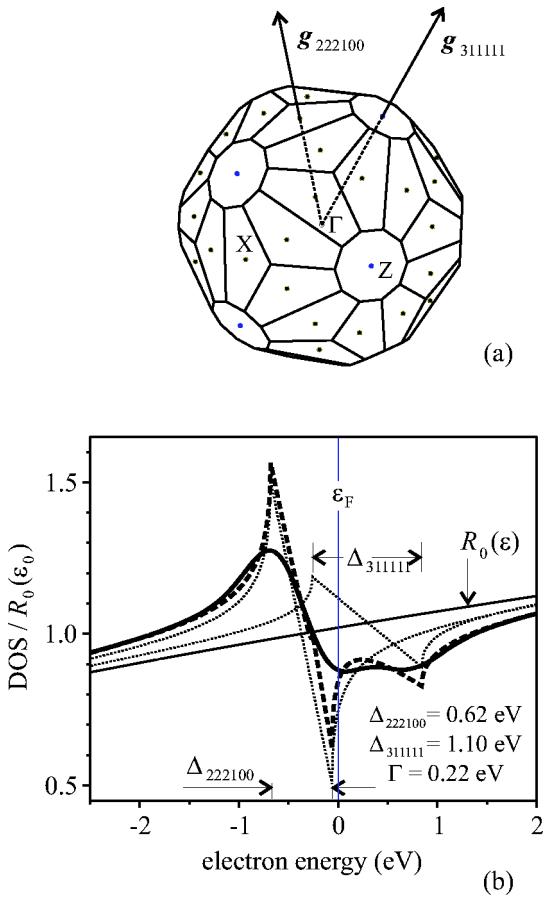


FIG. 5. The 222100 and 311111 Jones zone of icosahedral QC's and the scheme of the *i*-ZnMg(Y,Ho) density of states.

ing a linear dependence of the normal DOS in the vicinity of the Fermi level, $R_0(\varepsilon)=b+a(\varepsilon-\varepsilon_F)$. Results of these calculations are represented by full curves in Fig. 4 and, as clearly seen, nicely reproduce the experimental data. The best fit with the least-squares technique for the *i*-ZnMgY spectrum has been obtained at the following set of parameters: $\Delta_{222100}=0.63$ eV, $\Delta_{311111}=1.16$ eV, $\Delta\varepsilon_F=0.68$ eV, $\Gamma=0.23$ eV, and $s=0.1$ eV (where $\Delta\varepsilon_F$ is the position of the Fermi level with respect to the lower edge of the $\Delta_{222100}$ pseudogap). The analysis of the *i*-ZnMgHo spectrum yielded similar values: $\Delta_{222100}=0.61$ eV, $\Delta_{311111}=0.98$ eV, $\Delta\varepsilon_F=0.68$ eV, $\Gamma=0.22$ eV, and $s=0.09$ eV. Note, that the values derived for the instrumental energy resolution parameter, $s=0.1$ eV and $s=0.09$ eV, actually coincide with that determined from the analysis of the Fermi edge PE spectra of the reference gold sample, $s=0.1$ eV. This confirms the logical consistency of the model employed. The small difference in the two sets of $\Delta_{222100}$, $\Delta_{311111}$, $\Delta\varepsilon_F$, and $\Gamma$ parameters for *i*-ZnMgY and *i*-ZnMgHo is not essential, since this difference is within the error interval, $\approx 20\,\%$, which was estimated upon analyzing PE Fermi edge spectra, recorded from various specimens of a series of cleaved samples and at various photon energies within 20–60 eV interval.

The dashed curves in Fig. 4 present the calculated density of states (without broadening). As seen, the feature of the PE Fermi edge spectra at $\approx 0.7$ eV below the Fermi level is due first of all to the $\Delta_{222100}$ pseudogap, which is located below the Fermi level, and is affected by the lower edge of the $\Delta_{311111}$ pseudogap. The reconstructed *i*-ZnMg(Y,Ho) density of states is presented separately in Fig. 5(b), where dotted curves correspond to the partial contributions of the 222100 and 311111 Bragg planes, the dashed one is their superposition, and the full curve shows the broadened DOS.

The van Hove singularities of the density of states, which correspond to edges of the pseudogaps, are smoothed in the experimental PE spectra due to the finite instrumental energy resolution and the intrinsic broadening. The determined broadening parameter $\Gamma\approx 0.22$ eV corresponds to a lifetime of $\tau\approx 3\times 10^{-15}$ s, which is one order of magnitude shorter than the electron relaxation time in pure Zn and Mg metals at 77 K.

Although the analysis of a series of PE spectra in the vicinity of the Fermi level yielded actually the same parameters of the *i*-ZnMg(Y,Ho) DOS, the derived pseudogap $\Delta_{222100}$ and $\Delta_{311111}$ values most probably can be considered only as rough estimates. In fact, due to the high icosahedral symmetry the distortions of the isoenergetic surface at intersections with individual Bragg planes may essentially overlap, and in such a case the model of independent distortions employed here cannot be strictly justified. A reliable deter-

mination of the QC pseudopotentials would require PE measurements at ultrahigh energy resolution, for a more accurate model of the density of states, as well as independent experimental investigations, which would probe the energy spectrum above the Fermi level. Nevertheless, we would like to note that the value $\Delta_{311111} \approx 1.1$ eV, derived from the present PE data, is close to the 1.25 eV value of the *i*-ZnMgY pseudogap, evaluated from the analysis of *i*-ZnMgY optical spectra.[14]

Figure 5(b) presents the density of states reconstructed from PE data in the vicinity of Fermi level. To extrapolate the DOS to a whole valence band, one can use expression (3) assuming that the normal DOS is determined by the usual free-electron formula

$$R_0(\varepsilon) = \frac{1}{2\pi^2}\left(\frac{2m^*}{\hbar^2}\right)^{3/2}\sqrt{\varepsilon}. \tag{8}$$

Here $m^*$ is the effective DOS mass to be determined from the normalization condition[20]

$$n = \int_0^{\varepsilon_F} d\varepsilon\, R(\varepsilon), \tag{9}$$

where $n$ is the valence electron concentration, which yielded $m^* = 0.88\, m_0$ for *i*-ZnMgY. The fact that the effective DOS mass differs from the free-electron mass but is close to it, indicates that the DOS below the 222100 and 311111 pseudogaps is affected by quasiperiodic potential, but to a lesser extent than in the vicinity of the Fermi level.

The resulting *i*-ZnMgY VB density of states is presented in Fig. 6(a). One can compare the *i*-$Zn_{62}Mg_{29}Y_9$ QC DOS deduced within the present model with the DOS of $Zn_{65.22}Mg_{28.26}Y_{6.52}$ ($Zn_{60}Mg_{26}Y_6$) hexagonal approximant calculated by the linear muffin-tin orbital technique (Krajčí and Hafner,[3] Oshio and Ishii[4]). Since the atomic composition of the approximant is rather close to that of the ZnMgY QC, one can expect their DOS to be similar. The approximant DOS is presented in Figs. 6(b) and 6(c). The strong peak at the bottom of the valence band is due to the Zn 3*d* shallow core level. The valence band above it, as seen, is rather similar to that of ZnMgY QC deduced within the present model. The main differences are the spiky structure, which is absent in our model, and a comparatively wide DOS peak at ca. 2 eV above the Fermi level, which is due mainly to Y 4*d* derived states, as seen from the partial Y-DOS and *d*-DOS contributions, presented separately in Figs. 6(b) and 6(c). The fact that the reconstructed *i*-ZnMgY QC DOS is rather close to the DOS calculated from the first principles, without *a priori* assumption of the existence of the $\varepsilon(\boldsymbol{k})$ relation, is a sound support for the present model.

The *i*-ZnMgY DOS obtained can be used to evaluate the electronic specific heat, which, as known, is determined solely by the DOS value at the Fermi energy,

$$c_v = \gamma T, \quad \gamma = \frac{\pi^3}{3} R(\varepsilon_F) k_B^2. \tag{10}$$

The $R(\varepsilon_F) = 0.25\ (\text{eV atom})^{-1}$ value obtained within our model [Fig. 6(a)] corresponds to the Sommerfeld coefficient

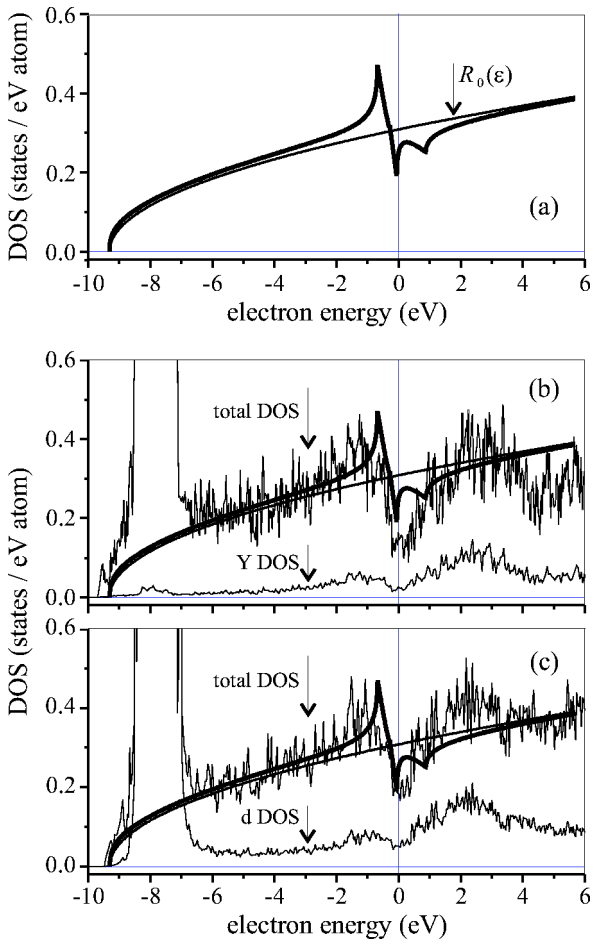


FIG. 6. The density of states of the icosahedral $Zn_{62}Mg_{29}Y_9$ quasicrystal (a) and its comparison with the DOS of the $Zn_{65.22}Mg_{28.26}Y_{6.52}$ hexagonal approximant (b) (Krajčí and Hafner, Ref. 3) and (c) (Oshio and Ishii, Ref. 4).

$\gamma = 0.59$ mJ/(mol $K^2$), which is very close to that measured on the *i*-ZnMgY polycrystalline samples, 0.63 mJ/(mol $K^2$),[15] and on the single-grain ones, 0.623 mJ/(mol $K^2$),[16] and 0.6 mJ/(mol $K^2$).[17]

Summarizing, the experimental low-temperature PE spectra of icosahedral ZnMgY and ZnMgHo quasicrystals indicate their simple metal-type valence bands at least down to ca. 5 eV below the Fermi level with a distinct pseudogap feature. The analysis performed suggests that the feature is due to van Hove singularities originating from intersections of the isoenergetic surface with the 222100 and 311111 families of Bragg planes. The reconstructed *i*-ZnMgY density of states agrees well with the main DOS features derived from the first-principles calculations for hexagonal ZnMgY approximants and reproduce the correct value of *i*-ZnMgY electronic specific-heat coefficient.

The model proposed does not exclude the possibility of the localized electron states in ZnMg(Y,Ho) quasicrystals. The eventual partial localization is indirectly indicated by the short lifetime of the electron states of the order of $10^{-15}$ s, as well as by the effective DOS mass reducing density of states below the Fermi level.

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[18] The spectral intensity background [dashed curves in Fig. 2(a)] has been approximated by the second-order polynomial, two coefficients of which have been determined to eliminate the background above the Fermi level, while the third one has been chosen such that the slope of the spectra below the pseudogap feature, at $\varepsilon_b < -2$eV, after the background subtraction should correspond to the slope of the $h\nu = 44$eV spectrum [Fig. 2(a)].

[19] The Fermi wave vectors $k_F = (3\pi^2 n)^{1/3}$ of ZnMg (Y, Ho) can be determined from the atomic compositions ($Zn_{62}Mg_{29}Y_9$ and $Zn_{65}Mg_{25}Ho_{10}$), the mass densities (5.18 g/cm$^3$ and 5.82 g/cm$^3$) of the quasicrystals, and the average valence of their atoms (2.09 and 2.10).

[20] Formula (4) contains the usual NFE approximation artefact at very small electron energies, $\varepsilon \sim N\Delta^2/16\varepsilon_0$, which is due to a violation of the perturbation procedure at the small energies. The artefact is not essential for the considered DOS features and was removed phenomenologically by multiplying the deviation function (4) by the $\varepsilon/(\varepsilon + N\Delta^2/16\varepsilon_0)$ factor.