

ISAACS – interactive structure analysis of amorphous and crystalline systems

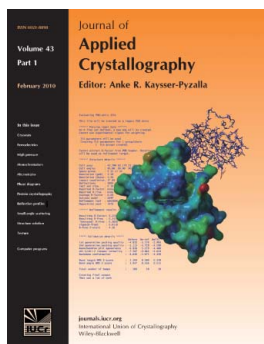
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ISAACS – interactive structure analysis of amorphous and crystalline systems

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ISAACS (interactive structure analysis of amorphous and crystalline systems) is a cross-platform program developed to analyze the structural characteristics of three-dimensional structure models built by computer simulations. The models may have any degree of periodicity (*i.e.* crystallinity) and local symmetry. The following structural information is computed from the models: total and partial radial distribution functions and structure factors for X-ray or neutron scattering, coordination numbers, bond-angle and near atomic neighbor distributions, bond-valence sums, ring statistics, and spherical harmonics invariants. The information may be visualized conveniently and stored for further use.

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1. Introduction

Detailed knowledge about the three-dimensional atomic scale structure is a prerequisite to understanding and predicting properties of materials. This knowledge is usually obtained by diffraction experiments followed by computer simulations aimed at building three-dimensional structure models that are consistent with the diffraction data and all other structure-relevant information known from alternative sources. However, once built the models are just a set of coordinate triples for several tens, in the case of crystalline materials, to several thousands, in the case of heavily disordered materials, of atoms. Consequently, the models need to be analyzed further so that important details of the atomic scale structure, such as distribution of first atomic neighbors, bond angles and local symmetry, are well understood. Unfortunately, few computer

programs exist that could perform the latter task. Here we describe a computer program named *ISAACS* (interactive structure analysis of amorphous and crystalline systems), designed as an intuitive and user-friendly tool box for carrying out detailed analyses of three-dimensional atomistic models of any degree of periodicity and local symmetry. The periodicity may range from the infinite periodicity of crystalline lattices to none, as with clusters. The symmetry may be as high as in cubic crystals and as low as that of melts.

2. Programming framework

ISAACS (Fig. 1) is developed in C and Fortran90 and uses GTK+ (GTK, 2009) for the graphical user interface (GUI). The C part of the code is used as a binding to wrap the GTK+ GUI over the Fortran90 core routines used for the calculations. Separating the *ISAACS* GUI from the Fortran90 routines makes the latter very easy to re-implement in other programs. Basically the Fortran90 routines are controlled by the GUI with minor exceptions, where GTK+ functions are called from Fortran90 routines to update a bar indicating the progress of calculations.

2.1. Supported platforms

The GTK+ library is a highly portable environment, which allows *ISAACS* to be a cross-platform software. Microsoft Windows (32 bits), Linux (32 and 64 bits) and Mac OS X (Intel-based) versions of the program are available.

2.2. The *ISAACS* project file format

ISAACS uses an intuitive format for a project file (see Fig. 2), which contains all the parameters needed to set up a calculation of

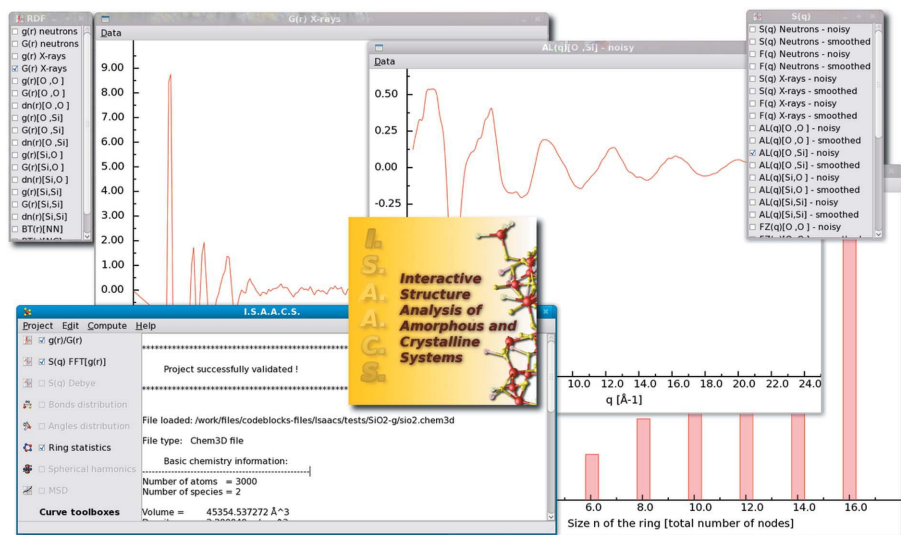


Figure 1
Visual overview of the *ISAACS* program.

structural characteristics of a three-dimensional model. The format of the file follows the XML coding (World Wide Web Consortium, 2009)

```
<?xml version='1.0' encoding='UTF-8'?>
<!-- ISAACS v1.0 XML file -->
<isaacs-xml>
<!-- Format and file of the configuration(s) -->
<data>
<type>Chem3D file</type>
<file>/home/leroux/Desktop/sio2.chem3d</file>
</data>
<!-- Chemistry information -->
<chemistry>
<atoms>3000</atoms>
<species number='2''>
<label id='0''>O</label>
<label id='1''>Si</label>
</species>
<element symbol='O''>
<name>Oxygen </name>
<z>8</z>
<mass>16.000000</mass>
<rad>0</rad>
<radius>0.660000</radius>
<nscatt>5.803000</nscatt>
<xscatt>8.000000</xscatt>
</element>
<element symbol='Si''>
<name>Silicon </name>
<z>14</z>
<mass>28.090000</mass>
<rad>0</rad>
<radius>1.110000</radius>
<nscatt>4.153000</nscatt>
<xscatt>14.000000</xscatt>
</element>
</chemistry>
<!-- Box information -->
<box>
<edges>
<a>35.662100</a>
<b>35.662100</b>
<c>35.662100</c>
</edges>
<angles>
<alpha>90.000000</alpha>
<beta>90.000000</beta>
<gamma>90.000000</gamma>
</angles>
<vectors>
<a.x>35.662100</a.x>
<a.y>0.000000</a.y>
<a.z>0.000000</a.z>
<b.x>0.000000</b.x>
<b.y>35.662100</b.y>
<b.z>0.000000</b.z>
<c.x>0.000000</c.x>
<c.y>0.000000</c.y>
<c.z>35.662100</c.z>
</vectors>
</box>
<!-- PBC information -->
<pbcs>
<apply>TRUE</apply>
<fractional>FALSE</fractional>
<fractype>0</fractype>
</pbcs>
<!-- Bond information -->
<cutoffs>
<total>2.184304</total>
<partials>
<O-O>2.808390</O-O>
<O-Si>2.184304</O-Si>
<Si-O>2.184304</Si-O>
<Si-Si>3.432477</Si-Si>
</partials>
</cutoffs>
<!-- Apply project -->
<project>TRUE</project>
</isaacs-xml>
```

Figure 2
Example of ISAACS project file (.ipf) in XML format, for glassy silica.

and allows the user to store detailed information about the model to be analyzed: chemical composition, the chemical and physical properties of each atomic species (e.g. atomic weight and X-ray/neutron scattering amplitudes), the size of the model box, the atomic coordinate type (e.g. Cartesian or fractional), time series properties, and a description of the bonding between atoms. The information is provided by the user and may be re-used/modified during ISAACS execution.

The .ipf file illustrated in Fig. 2 shows the parameters ISAACS needs to perform an analysis of the three-dimensional structure model of silica glass presented in Fig. 3.

In this particular example a file in 'chem3d' format (sio2.chem3d) comprising the coordinates of 3000 atoms from a three-dimensional structure model for silica glass is read by ISAACS. The corresponding atomic number Z ($Z = 8$ for oxygen and $Z = 14$ silicon), atomic mass, radius, and neutron and X-ray scattering amplitude are assigned to each of the atoms from the model. The values of these parameters for all atomic species from the periodic table are stored in ISAACS but may be modified by the user if needed. The size of the model box in terms of box edge lengths a , b and c and angles α , β and γ are provided by the user. Here the box is cubic and its edge is 35.662 Å in length (see Fig. 2). ISAACS then generates a transformation matrix of vectors to be used as an internal coordinate system. Finally, the bond-length distances (e.g. O—O, Si—O and Si—Si) are specified by the user.

3. Features

The main interface of the ISAACS program (Fig. 4a) gives access to the following menus:

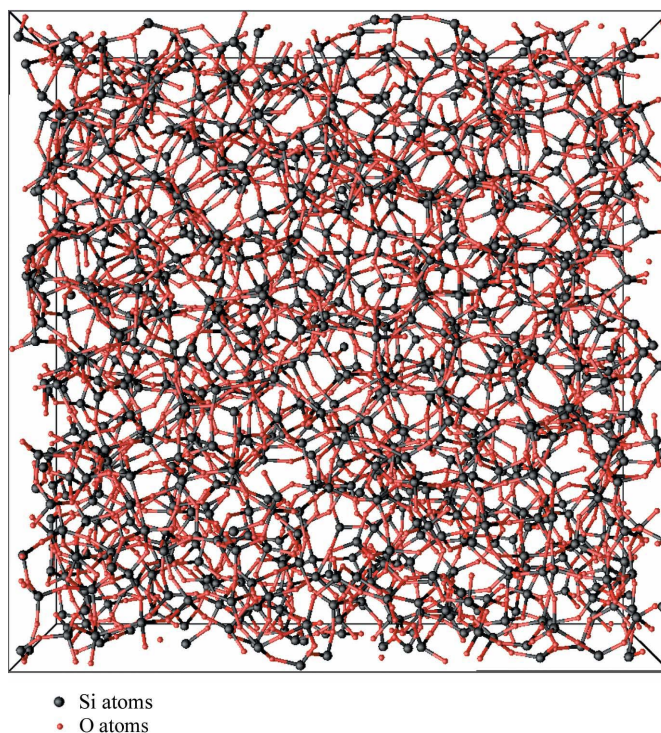


Figure 3
Three-dimensional structure model of silica glass generated by reverse Monte Carlo simulations guided by synchrotron X-ray radiation data. The glass is a continuous random network of Si and O atoms sitting, respectively, at the centers and vertices of only corner sharing tetrahedral units.

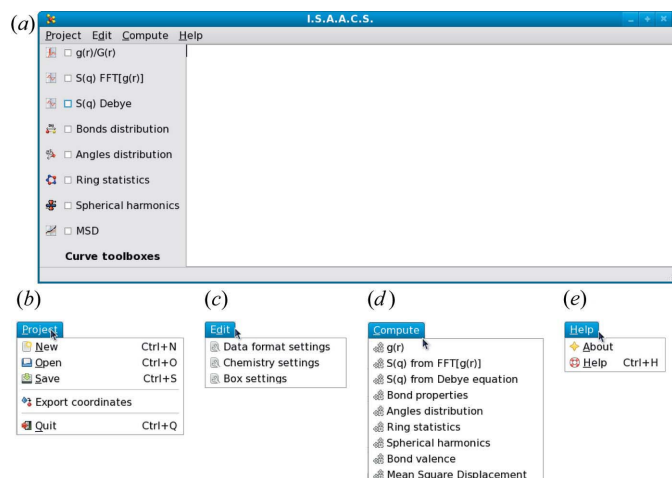


Figure 4

(a) Main interface of the *ISAACS* program, with the (b) ‘Project’, (c) ‘Edit’, (d) ‘Compute’ and (e) ‘Help’ menus.

(i) The ‘Project’ menu (Fig. 4b) is used to read and write .ipf files (Fig. 2) as well as to import/export coordinates of atoms from the analyzed structure models (§3.1).

(ii) The ‘Edit’ menu (Fig. 4c) is used to define the properties of the structural characteristics of a system to be studied.

(iii) The ‘Compute’ menu (Fig. 4d) is used to run the calculations (§3.2).

(iv) The ‘Help’ menu (Fig. 4e) is used to access the documentation provided to help users.

3.1. Data inputs and outputs

The current version of *ISAACS* can import three-dimensional structure models in four different formats:

- (1) XYZ (Minnesota Supercomputer Center, 1993)
- (2) multiple XYZ (Minnesota Supercomputer Center, 1993)
- (3) Chem3D (CambridgeSoft, 2009)
- (4) PDB (Protein Data Bank; Berman *et al.*, 2000)

The coordinates of atoms from a three-dimensional model can be in any of the widely used formats listed below:

- (1) Cartesian
- (2) Fractional with the center of the model box at (0, 0, 0)
- (3) Fractional with one of the corners of the model box at (0, 0, 0)

ISAACS can output an already imported three-dimensional model in XYZ or multiple XYZ structure formats (Minnesota Supercomputer Center, 1993) in any of the atomic coordinate formats (1)–(3).

3.2. What can be computed by *ISAACS*?

ISAACS can compute the following important structural characteristics of a three-dimensional structure model:

(a) Radial distribution functions $g(r)$ (RDFs) (Allen & Tildesley, 1987), including (i) total RDFs for neutrons and X-rays; (ii) partial RDFs; and (iii) Bathia–Thornton RDFs (Salmon, 2007).

(b) Structure factors $S(q)$ (Dove *et al.*, 2002) including¹ (i) total structure factors $S(q)$ for neutrons and X-rays; (ii) Faber–Ziman (Faber & Ziman, 1965) partial $S(q)$; (iii) Ashcroft–Langreth

¹ Structure factors can be computed by (i) Fourier transformation of the radial distribution functions and/or (ii) using the Debye formalism (Dove *et al.*, 2002).

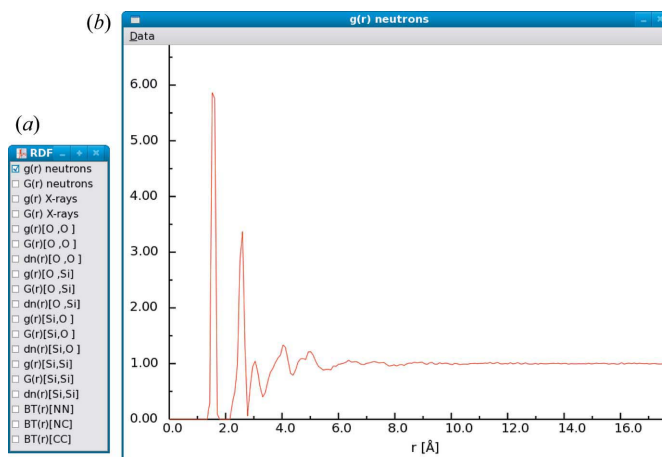


Figure 5

Results of the calculation of the radial distribution functions for a 3000-atom model of silica glass by the *ISAACS* program: the interaction box (a) displays the corresponding *ISAACS* menu options, while (b) shows the computed RDF in the case of neutron scattering. The RDF exhibits a first sharp peak positioned at 1.6 Å, which is the Si–O distance in well defined Si(O)₄ tetrahedral units. The second RDF peak, at 2.4 Å, reflects the correlations between O atoms sitting on the vertices of those tetrahedra.

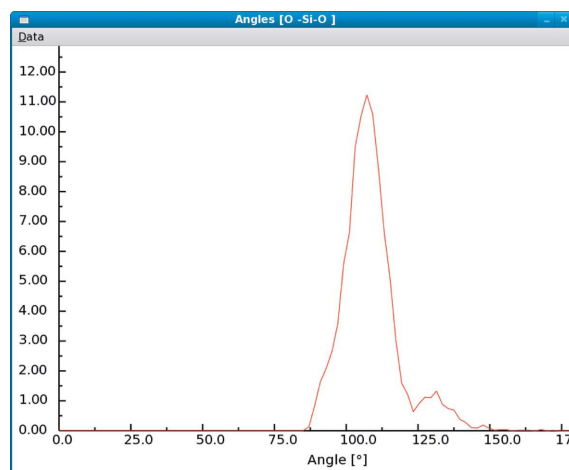


Figure 6

Results from the calculation of the angle distribution for a 3000-atom model of silica glass by the *ISAACS* program: the figure represents the distribution of the bond angles (O–Si–O) as computed and displayed by *ISAACS*. In SiO₂ glass the O–Si–O angle distribution peaks at about 109° as may be expected for tetrahedral Si(O)₄ units.

(Ashcroft & Langreth, 1967*a,b*, 1968) partial $S(q)$; and (iv) Bathia–Thornton (Bhatia & Thornton, 1970) partial $S(q)$.

In *ISAACS* the existence of a bond between particular atomic species is determined by the user, who may supply appropriate bond-length distances (see Fig. 2). An estimate for the bond-length distances in a structure model may be made by looking at the first maxima/minima of the respective partial atomic distribution distances computed by *ISAACS*.

When atomic bonds in a model are defined properly other structural characteristics can be evaluated, as follows:

(c) Interatomic bond properties: (i) coordination numbers; (ii) atomic near neighbor distribution; (iii) fraction of links between tetrahedra; (iv) fraction of tetrahedral units; and (v) bond-length distribution for the first coordination sphere.

(d) Distribution of bond angles.



Figure 7

Results of the calculation of the spherical harmonics as local order parameters for a 3000-atom model of silica glass by the *ISAACS* program: (a) average Q_i computed for the $\text{Si}(\text{O}_4)$ environments (distorted tetrahedra) in glassy SiO_2 , compared with (b) average Q_i computed for the $\text{Si}(\text{O}_4)$ environments (ideal tetrahedra) in crystalline quartz- α . The two sets of Q_i values follow a similar trend, showing the similarity between the structural units in crystalline and glassy SiO_2 . The differences are mostly quantitative and are due to the fact that the Si–O tetrahedra are somewhat distorted in the glass.

(e) Distribution of dihedral angles.

(f) Ring statistics, according to several definitions: (i) all closed paths (no rules); (ii) King's (1967) rings; (iii) shortest paths (Guttman, 1990; Franzblau, 1991); (iv) primitive rings (Goetzke & Klein, 1991; Yuan & Cormack, 2002) or irreducible (Wooten, 2002); and (v) strong rings (Goetzke & Klein, 1991; Yuan & Cormack, 2002). Also included are options for (vi) the possibility to look only for *ABAB* rings and (vii) the possibility to exclude rings with homopolar bonds (*A–A* or *B–B*) from the analysis. Ring statistics are presented according to the *RINGS* method (Le Roux & Jund, 2010).

(g) Spherical harmonics invariant, Q_i , as local symmetry identifiers (Steinhardt *et al.*, 1983): (i) average Q_i for each chemical species and (ii) average Q_i for a user-specified structural unit.

(h) Bond-valence sums (CCP14, 2006; Brown & Altermatt, 1985; Brown & Shannon, 1973): (i) average bond valence for each chemical species and (ii) average bond valence for a user-specified structural unit.

ISAACS also computes the mean square displacement (MSD) of atoms, a dynamic property widely used in the analysis of molecular

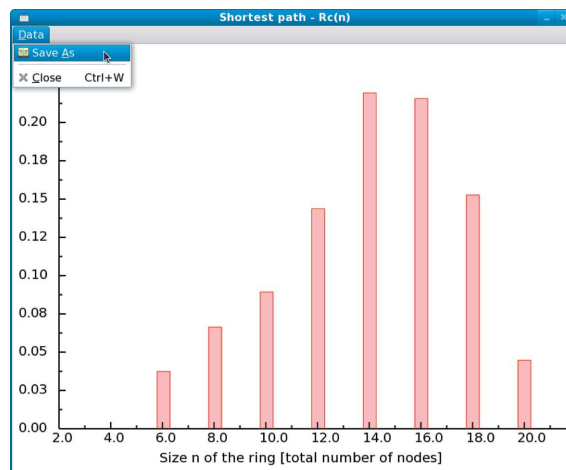


Figure 8

Ring statistics for silica glass as computed by *ISAACS*. These results present the average number of rings per atom in the simulation box for a maximum ring size fixed to 20 atoms. Note that if the number of rings is normalized per $\text{Si}(\text{O}_4)$ unit and not per atom the 'ring size' will drop by a factor of two, *i.e.* the distribution will peak around rings made of seven $\text{Si}(\text{O}_4)$ units.

dynamics simulations. In that case *ISAACS* can compute the following quantities:

- (i) MSD per atomic species.²
- (j) Directional MSD (x, y, z, xy, xz, yz).²
- (k) Drift of the center of mass (x, y, z).
- (l) Correction of the drift of the center of mass (x, y, z).

3.3. General features of the program

A set of structural characteristics for a three-dimensional model of silica glass computed by *ISAACS* is shown (see Figs. 5–8), as an example of the program output. The model has been constructed by reverse Monte Carlo simulations guided by high-energy X-ray diffraction data for silica glass (Petkov *et al.*, 2000). It consists of 2000 O and 1000 Si atoms inside a box of dimensions 35.6621 Å. To compute each of the characteristics shown below the relevant dialog/menu boxes have been used to set up the values of the program parameters required to run a structure analysis. For example, Fig. 9 presents the dialog boxes used to control the calculations of the radial distribution function (Fig. 9a) and the ring statistics (Fig. 9b) for silica glass. Once the three-dimensional structure is read and validated by *ISAACS* the only parameter required to compute the RDF is the number of steps in real space, δr (Fig. 9a). In the second case (Fig. 9b) a few more control parameters (the definition of a ring, the chemical species used to initiate the search, the maximum size of a ring, the maximum number of rings per node and a description of the chemical bonds) are needed.

ISAACS requires users to supply control parameters to guide the computation sequence. Information about these parameters can be obtained through a help button in the dialog box (Fig. 9) or through the help menu of the main window of the program (Fig. 4e).

3.4. Visualization of the results of the calculations

When a particular structural characteristic is computed it can be directly displayed in the main *ISAACS* windows (Fig. 4). In addition the visualization mode of most of the computed characteristics can be

² Computation performed with a correlation of all the properties as well as without correlation (step by step).

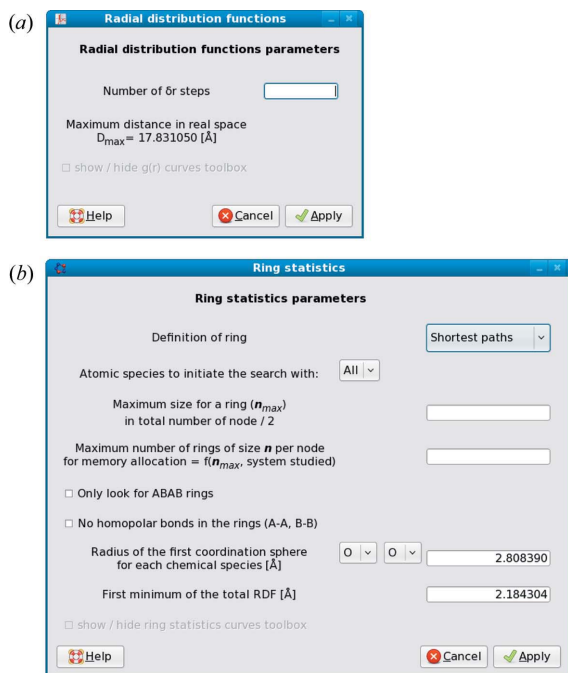


Figure 9
Dialog boxes controlling (a) the calculation of radial distribution functions and (b) ring statistics in the ISAACS program.

controlled *via* interactive menus such as the one presented in Fig. 5(a). When a button in an interaction menu is activated (Fig. 5a) the corresponding result is instantaneously displayed as a smooth curve or a histogram (Figs. 5b, 6, 7 and 8) depending on the nature of the computed structural characteristic.

3.5. Saving the data

Results computed by ISAACS can be easily saved using the standard copy and paste method (for the results presented in the main ISAACS window) or using the 'Data' menu (Fig. 8). The user can also export data either in a raw ASCII format (simple two-column file with x and y) or in the Xmgrace format, which can be used immediately in the Grace plotting tool (<http://plasma-gate.weizmann.ac.il/Grace/>) for a further analysis.

Fig. 10 shows the data of Fig. 8 as exported and plotted by the Xmgrace export filter of ISAACS.

4. Distribution and documentation

The ISAACS program is licensed under the terms of the GNU General Public Licence version 3 (Free Software Foundation, 2007). ISAACS binaries and sources are available for download from the following web sites: <http://www.phy.cmich.edu/people/petkov/isaacs/> and <http://isaacs.sourceforge.net/>.

The web sites dedicated to the ISAACS program also provide a theoretical background of the structural analysis, model examples, an

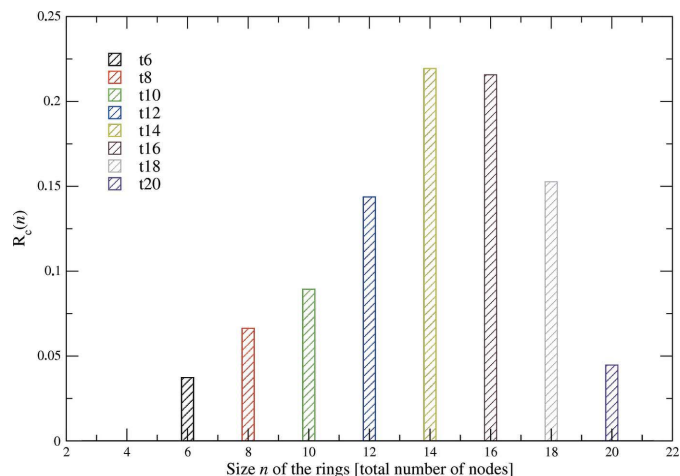


Figure 10
Illustration of the utilization of the Xmgrace export filter in the ISAACS program.

82 page user manual and online help on the utilization of the program.

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