Experimental Investigation of Low Dimensional Spin System in Metal Oxides

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Abstract
Nano particles of SrCrO\textsubscript{4} were manufactured by sol-gel technique. The crystal part of SrCrO\textsubscript{4} is monoclinic having space group P21/n. We calcined it at 950°C temperature. Its c and a lattice parameter are 6.77 and 7.08 which is very close to the reported ones. Ba and Ca-doped SrCrO\textsubscript{4} were also synthesized by sol-gel method by various concentrations for x= 0.2, 0.4, 0.6 and 0.8. Doping samples also sintered at 950°C for 2 hours in order to obtain fine powder. Different characterization techniques such as XRD, Ultra-violet-Spectroscopy, PL Spectroscopy and FTIR Spectroscopy were used to analyze SrCrO\textsubscript{4}, Ba and Ca-doped SrCrO\textsubscript{4}. XRD tells us about the crystal size and dislocation density of samples. The value of 2θ for the XRD patterns is ranging from 5 to 95. Four peaks are observed in the UV-spectra of SrCrO\textsubscript{4} which occur at 350nm, 380nm, 700nm and 750nm. The UV band gap of SrCrO\textsubscript{4} is 3.25 eV. In PL spectra, two peaks are observed one at 480nm and other at 410nm. At 480nm, the energy of emitted photons is 2.5eV, while at 410nm the emitted photons has energy of 3ev. In the FTIR analysis, the core modes frequently showed by CrO\textsubscript{4} unit are symmetric stretching bond (υ1 (A1)), symmetric bending mode (υ2 (E)), asymmetric stretching mode (υ3 (F2)) and asymmetric bending mode (υ4 (F2)).

Keywords
Heusler compounds, Energy gaps, Reflectivity, Interband transitions.

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Condensed Matter Physics

The different fields of Physics, like metallurgy, magnetism, crystallography, and elasticity that were dealing with the different aspects of solid matter were considered as the self-determining areas of science, in the age of 20th century. During the era of 1940, these fields were combined with many other different fields and then they were collectively named as “Solid State Physics” which two years later was then magnified to include the study of physical properties of liquids and then this field was known as Condensed Matter Physics. This is the largest subfields of physics. According to the estimation of American Physicist, the Condense Matter Physics is closely related to the field of material science (Kohn, 1999).

Strongly Correlated Electron (SCE) Systems

In Condensed Matter Physics, the most intriguing phenomena is “Strongly correlated electron (SCE) system”. We can easily understand the physical properties of various materials like simple metals, insulators and some semiconductors through the modern-solid state physics. The properties of materials having open d and f electron shells, where electrons lodge restricted orbitals are difficult to understand. Electrons in transition metals like Iron, Vanadium and their oxides involvement strong cumbic dislike because of the longitudinal limitations in their orbitals. Such type of strongly interrelating or “correlated” electrons which are generated by other electrons cannot be explained by means of static mean field. The effect of correlations on the characteristics of materials is often reflective. The phenomena of interchange between interior grades of autonomy, d and f electrons, orbital moment, charge and spin can show a whole venture of unusual ordering at low temperature. Thus, because of this SCE the system becomes intensely delicate to small changes in outside parameters, just like pressure, fixing and temperature. Some SCE materials show a great change in an applied magnetic field; others exhibit a large thermoelectric response, an effect labeled as colossal magneto-resistance (Kotliar & Vollhardt, 2004).

In the SCE materials, the migration of one electron perturbed the movements and positions of all the other electrons due to the long-range coulomb interaction. On the basis of this interpretation, one would conclude that all materials exhibit the phenomena of strong electronic correlations. Contrarily, in the case of pure ionic systems, the electrons are preoccupied in the atomic nucleus of nearby atom. However, in an ideal metallic systems, the other conduction electrons display the long-range Coulomb interaction. Consequently, as electronic correlations are present in both metallic and ionic systems just like as in Magnetism and Lead. Many simple models are helpful in explaining the main properties of these systems at that point where electronic correlations are taken into account or either completely neglected only in low order approximations. These approximations break down and completely new functionalities and phenomena can appear in highly correlated electronic systems. These so-called emergent phenomena among the electrons, between the electrons and the lattice cannot be interpreted through local interactions (Laughlin & Pines, 2000).

Introduction

In this section, we discussed about the techniques and instruments which are used for the synthesis and characterization of SrCrO4 nanoparticles. Later we will explain different techniques of characterization such as UV-VIS Spectroscopy, XRD, Photoluminescence Spectroscopy and Fourier-Transform Infrared Spectroscopy.

Different methods of Sample Synthesis
Synthesis of sample is a process in which an exemplary part of material is interpreted from a considerable amount and then arranged for the analysis. Sample synthesis and sampling both have same meaning and in the field of analytical chemistry it perform a significant role.

To obtain the homogeneous and single phase of strontium chromate SrCrO$_4$ nanoparticles, different methods may be applied. Different methods which are used for the synthesis of SrCrO$_4$ nanoparticles are listed below.

i. Co-precipitation Method
ii. Solution-phase Method
iii. Sol-Gel Method
iv. Bio-template Method
v. Combustion Method

**Sol-Gel Method**

In order to synthesize new materials, sol-gel method provides a new path. Sol-gel method during the synthesis of samples carefully controlled the entire reaction. Solid materials are produced from small molecules by sol-gel procedure in material science. The sol-gel method may be defined as, “molecules when react with each other in a liquid via poly condensation process, an oxide network is formed”. In nuclear industry due to the basic requirement of new synthesis methods, a sol-gel method was introduced in 1960’s. This method takes place at a low sintering temperature.

Materials having different shapes for example thin films, porous structures and dense powders may be synthesized with the help of sol-gel method. The sol-gel technique is a saturated method. For the synthesis of sample, the material first dissolved in distilled water to create liquefies part and then we return back into an extremely rigid form. In the sol-gel technique, the interrelated substances and tiny particles are arranged by a continuation process. Different fields of science such as chemical engineering and material science also utilized this technique.

The method of sol-gel in order to synthesize the sample consists of following steps:

Firstly, for the preparation of a solution all the calculated amount of chemicals are fused in de-ionized water.

Secondly, the solution is constantly agitating at low temperature on the hot plate and also at a definite point retain its PH.

Thirdly, by constant heating the particles which are in the solution form were converted into gel.

Lastly, by a very suitable heat treatment the gel form is finally converted into a fine powder.

**Uses of Sol-Gel Technique**

The most important advantage of sol-gel way is the preparation of organic-inorganic hybrids. Materials which are synthesized by this method are highly purified. As the sol-gel process starts from the solution, due to this it is quite versatile. Materials with different microstructures and shapes can be manufactured. Refractory materials are prepared by sol-gel method as compared to the conventional (square) method of melt-quenching and sintering at lower temperatures. The fabrication of silica aerogels is possible with the help of this method. Thermal insulation of these materials is very high. Mechanical properties of materials such as hardness and strength can be improved by sol-gel method. The optical filaments as well as intractable ceramics may also be achieved by modifying the thickness from this mode.
Composition of Sample

The progression of sol-gel method largely composed of chlorides, carbonates and nitrate materials. As our work is on nitrates, so for the synthesis of strontium chromate SrCrO$_4$, Ba and Ca-doped SrCrO$_4$ we used Strontium Nitrate Sr(NO$_3$)$_2$, Barium Nitrate Ba, Calcium nitrate and Chromium Nitrate Cr(NO$_3$)$_2$. For the formation of sample, the desired calculations of chemicals were done and in the de-ionized water the solution of these nitrates was formed.

Ratio of Chemicals:

Parent Sample SrCrO$_4$

Sr(NO$_3$)$_2$ = 3.701 g/20ml ethanol
Cr(NO$_3$)$_2$ = 7g/20ml ethanol
Citric acid = 7.35/20ml distilled water

Ba-doped SrCrO$_4$

For sample 1, x= 0.2
Cr(NO$_3$)$_2$ = 7g/20ml ethanol
Sr(NO$_3$)$_2$ = 2.827g/20ml ethanol
Ba(NO$_3$)$_2$ = 0.87g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

For sample 2, x= 0.4
Cr(NO$_3$)$_2$ = 7g/20ml ethanol
Sr(NO$_3$)$_2$ = 2.02g/20ml ethanol
Ba(NO$_3$)$_2$ = 1.67g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

For sample 3, x= 0.6
Cr(NO$_3$)$_2$ = 7g/20ml ethanol
Sr(NO$_3$)$_2$ = 1.29g/20ml ethanol
Ba(NO$_3$)$_2$ = 2.40g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

For sample 4, x=0.8
Cr(NO$_3$)$_2$ =7g/20ml ethanol
Sr(NO$_3$)$_2$ = 0.62g/20ml ethanol
Ba(NO$_3$)$_2$ = 3.07g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

Ca-doped SrCrO$_4$

For sample 1, x= 0.2
Cr(NO$_3$)$_2$ = 7g/20ml ethanol
Sr (NO₃)₂ = 2.89g/20ml ethanol
Ca (NO₃)₂ = 0.807g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

For sample 2, x= 0.4
Cr (NO₃)₂ = 7g/20ml ethanol
Sr (NO₃)₂ = 2.12g/20ml ethanol
Ca (NO₃)₂ = 1.57g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

For sample 3, x= 0.6
Cr (NO₃)₂ = 7g/20ml ethanol
Sr (NO₃)₂ = 1.38g/20ml ethanol
Ca (NO₃)₂ = 2.31g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

For sample 4, x=0.8
Cr (NO₃)₂ =7g/20ml ethanol
Sr (NO₃)₂ = 0.67g/20ml ethanol
Ca (NO₃)₂ = 3.02g/20ml ethanol
Citric acid = 7.35g/20ml distilled water

Results and Discussions

XRD Analysis

As XRD analysis gives us the information about the crystal structure and phase of a sample. The XRD arrangements of SrCrO₄ is displayed in figure 1. The value of 2θ for the XRD patterns is ranging from 5 to 95. So, the main peaks of our XRD results at 950°C are 24.03, 25, 27.24, 29.58, 46.01, 46.32 and 49.29. We obtain the single phase of our sample at 950°C temperature. The crystal structure of our sample is monoclinic which showed that sample attained had a high purity. Its space group is P2₁/n. We obtain the (114) phase with reference code 00-015-0368 of strontium chromate. The c and a lattice parameter of samples which analyzed at 950°C are 6.77 and 7.08 respectively. These c and a lattice parameter of our sample are closely related with the reported ones. Its ratio c/a is 0.95.
Bragg’s law:

\[ n\lambda = 2d\sin\theta \]

With the help of this equation, we can easily calculate the size of a particle. In this equation, \( n \) shows an number, \( \lambda \) stand for wavelength, \( d \) shows the inter-planar spacing and \( \theta \) is the incident angle between incident and scattered ray.

We can also calculate the crystal size of a particle by using the formula which is given below:

\[ D = \frac{K\lambda}{\beta \cos} \]

**Figure 1.** XRD pattern of SrCrO\(_4\)

**Figure 2.** XRD patterns of Sr\(_{1-x}\)Ba\(_x\)CrO\(_4\) with conc. \( x = 0.2, 0.4, 0.6, 0.8 \).
The XRD patterns of Ba-doped SrCrO$_4$ at temperature 950°C with various attentions $x=0.2, 0.4, 0.6, 0.8$ is shown in figure 2. In the figure we observed that at lower concentration $x=0.2$, we have very sharp and intense peaks. But as we start increasing the concentration of a sample, some peaks start to suppressed and finally they combine to give a single peak in the end just as in the case of $x=0.8$. This shows that with the increase of concentration of a sample, its intensity starts decreasing. The $a$-lattice parameters of Ba-doped SrCrO$_4$ at $x=0.2, 0.4, 0.6, 0.8$ are 7.67, 7.67, 7.65 and 7.62. The $c$-lattice parameters of Sr$_{1-x}$Ba$_x$CrO$_4$ at $x=0.2, 0.4, 0.6, 0.8$ are 10.39, 10.39, 10.1, 10.46. Its ratio $c/a$ are 1.35, 1.35, 1.32 and 1.37.

![XRD patterns of SrCrO$_4$ and Ba-doped SrCrO$_4$](image)

**Figure 3.** XRD patterns of SrCrO$_4$ and Ba-doped SrCrO$_4$

The XRD patterns of our parent sample SrCrO$_4$ is compared with the XRD pattern of Sr$_{1-x}$Ba$_x$CrO$_4$ at a temperature of 950°C. In figure 3. It can be undoubtedly detected that parent sample SrCrO$_4$ has very sharp peaks. But when it is doped with barium at different absorptions $x=0.2, 0.4, 0.6, 0.8$ its intensity starts to decrease. And at $x=0.8$, the intense peaks of Ba-doped SrCrO$_4$ suppressed. The $c$ and $a$ lattice parameters of Sr$_{1-x}$Ba$_x$CrO$_4$ is increased as compared with the lattice parameters of SrCrO$_4$ which is shown in table 1.

**Ca-doped SrCrO$_4$**
Figure 4: XRD patterns of Sr$_{1-x}$Ca$_x$CrO$_4$ with conc. x = 0.2, 0.4, 0.6, 0.8.

The XRD patterns of Ca-doped SrCrO$_4$ at 950°C temperature with concentrations x = 0.2, 0.4, 0.6, 0.8 is shown in figure 4. In this figure we noticed that at lower concentration x = 0.2, we have very intense peaks. But as we start increasing the concentration of a sample, most of the peaks starts to compressed and finally they combined with each other to give a single peak. In case of x = 0.8, the graph having very low peaks and its shows linearity. This shows that intensity decreases in our doping sample as we move from lower to higher concentration. The a-lattice parameters of Ca-doped SrCrO$_4$ at x = 0.2, 0.4, 0.6, 0.8 are 7.08, 7.08, 8.92, 10.00. The c-lattice parameters of Sr$_{1-x}$Ca$_x$CrO$_4$ at x = 0.2, 0.4, 0.6, 0.8 are 6.755, 6.78, 7.59, 5.8. Its ratio c/a are 0.95, 0.95, 0.85 and 0.58.
Figure 5. XRD patterns of SrCrO₄ and Ca-doped SrCrO₄

The XRD pattern of our parent sample SrCrO₄ is compared with the XRD pattern of Sr₁₋ₓCaₓCrO₄ at a temperature of 950°C. In figure 5, we observed that SrCrO₄ has very sharp peaks. But when it is doped with calcium at dissimilar concentrations x= 0.2, 0.4, 0.6, 0.8 its intensity start reducing. At x= 0.8, the sharp peaks of Ca-doped SrCrO₄ almost diminishes and the graph moves towards linearity. A lattice parameter of Sr₁₋ₓCaₓCrO₄ is increased and its c-lattice parameter is decreased as compared with the lattice parameters of SrCrO₄ which is shown in table 2.

| Samples   | a-LP | c-LP  | c/a  |
|-----------|------|-------|------|
| SrCrO₄    | 7.08 | 6.77  | 0.95 |
| Ba-0.2%   | 7.67 | 10.39 | 1.35 |
| Ba-0.4%   | 7.67 | 10.39 | 1.35 |
| Ba-0.6%   | 7.65 | 10.1  | 1.32 |
| Ba-0.8%   | 7.62 | 10.46 | 1.37 |

Table 1. The a and c-LP of SrCrO₄ and Ba-doped SrCrO₄
### Table 2. The a and c-LP of SrCrO$_4$ and Ca-doped SrCrO$_4$

| Samples     | a-LP (nm) | c-LP (nm) | c/a   |
|-------------|-----------|-----------|-------|
| SrCrO$_4$   | 7.08      | 6.77      | 0.95  |
| Ca-0.2%     | 7.08      | 6.75      | 0.95  |
| Ca-0.4%     | 7.08      | 6.78      | 0.95  |
| Ca-0.6%     | 8.92      | 7.59      | 0.85  |
| Ca-0.8%     | 10.00     | 5.8       | 0.58  |

### Table 3: Crystal size and dislocation density of SrCrO$_4$ and Ba-doped SrCrO$_4$

| Samples     | Crystal size (D) nm | Dislocation density $\delta$ (nm)$^2$ |
|-------------|---------------------|--------------------------------------|
| SrCrO$_4$   | 13.711              | 0.0053                               |
| Ba-0.2%     | 11.544              | 0.0075                               |
| Ba-0.4%     | 10.570              | 0.0089                               |
| Ba-0.6%     | 11.709              | 0.0072                               |
| Ba-0.8%     | 30.24               | 0.0010                               |

### Table 4. Crystal size and dislocation density of SrCrO$_4$ and Ca-doped SrCrO$_4$

| Samples     | Crystal size (D) nm | Dislocation density $\delta$ (nm)$^2$ |
|-------------|---------------------|--------------------------------------|
| SrCrO$_4$   | 13.711              | 0.0053                               |
| Ca-0.2%     | 18.275              | 0.0029                               |
| Ca-0.4%     | 19.137              | 0.0027                               |
| Ca-0.6%     | 23.073              | 0.0187                               |
| Ca-0.8%     | 9.129               | 0.0119                               |

**UV-VIS Analysis:**

In this type of spectroscopy, photons of wavelength lying in the UV-Visible region, fall on the material under observation. From valence to the conduction band photon having energy equal or greater will be absorbed during transform electron, thus creating electron in the conduction band.
and hole in the valence band. Electron may occupy different sub-bands in the transmission band depending on the energy of photons absorbed. When photons are absorbed, the spectra can be obtained that will show different peaks. The peaks may show different band gap energy, less, equal or larger than the band gap of the material.

![Absorbance vs Wavelength](image)

**Figure 6.** shows the UV spectra of SrCrO$_4$ at 950°C having wavelength range of 200-800nm. Four peaks are observed in this graph which occur at 350nm, 380nm, 700nm and 750nm.

![UV band gap of SrCrO$_4$](image)

**Figure 7.** UV band gap of SrCrO$_4$

In figure 7. The UV band gap curve of our SrCrO$_4$ is shown. Spectra of the sample calcined at 950°C shows that the band gap shifts towards higher energy i.e 3.25 eV. As the calcination temperature
becomes high the band gap tunes towards actual band gap of the material that is due to removal of impurities.

**PL Analysis:**

In photoluminescence (PL) electron once excited to the conduction band will never be there it will de-excite emitting photons of certain energy. The energy of emitted photons is plotted vs intensity.

![PL spectra of SrCrO₄ nanoparticles](image)

**Figure 8.** PL spectra of SrCrO₄ nanoparticles

In our case the spectra shown in figure 4.8. In each spectra two peaks are observed one at 480 nm and other at 410 nm. At 480 nm the energy of emitted photons is 2.5 eV which is less than band gap of the material and occurs due to sub-bands that is being introduced by oxygen vacancies and other defects while at 410 nm the emitted photons has energy of 3 eV that occur due to band-band transition of electron.

**FTIR Analysis**

Figure 9. Shows FTIR spectrum of SrCrO₄ annealed at 950°C. The spectrum was developed from 4000-450 cm⁻¹, but only the variety of 400-1200 cm⁻¹ was related to the current conversation.
The interior modes frequently displayed by CrO$_4$ unit are symmetric stretching bond (υ$_1$ (A$_1$)), symmetric bending mode (υ$_2$ (E)), asymmetric stretching mode (υ$_3$ (F$_2$)) and asymmetric bending mode (υ$_4$ (F$_2$)). The band in this range were recorded as shown in figure 9. The bands indexed at 901, 885 and 870 cm$^{-1}$ correspond to the Cr-O asymmetric stretching mode (υ$_3$ (F$_2$)) and the band centered at 834 cm$^{-1}$ represents the Cr-O symmetric stretching mode (υ$_1$ (A$_1$)). The O-Cr-O symmetric and asymmetric bending modes which are usually found below 450 cm$^{-1}$ for this compound are not presented in the figure due to the limitation of the scan range of the instrument.

Fig 9 shows the ftir spectra of SrCrO$_4$ and fig 10 shows ftir spectra of Ba doped SrCrO$_4$ at different concentration rate. Ba is doped with 0.2, 0.4 and 0.6 doping concentrations and their peaks lie in
the 500-700 and 800-1000 range. In 0.2 concentration some peaks are suppressed but as we go increasing amount of dopant these peaks are going more enhanced.

In fig 11. comparison of both SrCrO$_4$ with the Ba doped with different concentrations FTIR spectra is given. In comparison we can see that after doping these peaks are increasing in the downward direction and some peaks expands.

![FTIR spectra of SrCrO$_4$ and Ba-doped SrCrO$_4$](image)

**Fig 11.** is the ftir spectra of Ca-doped SrCrO$_4$ and fig 13 is the ftir spectra of SrCrO$_4$ with comparison of Ca doped sample. And after doping in Ca these peaks are also going in downward and these are lie in the region where SrCrO$_4$ lie.

Three bands ranges high frequency, mid frequency and low frequency ranges are 670-570, 480-350 and at 190 cm$^{-1}$ respectively. IR mode are similar to these.
Figure 12. FTIR analysis of Ca-doped SrCrO$_4$

Figure 13. FTIR analysis of SrCrO$_4$ and Ca-doped SrCrO$_4$
High-frequency band:
High frequency band region from 570-670 cm\(^{-1}\). This band is mainly due to the Cr-O-Cr asymmetric stretching. This mode is inversely proportional to bond length of Cr-O. The Cr-O stretching band appear at 630 cm\(^{-1}\).

Low-frequency Band:
The moderately strong mid-frequency band is again a super-position of many modes and is spread from 350-480 cm\(^{-1}\). These modes arise due to the deformation. O-Cr-O modes are also reported here.

Conclusions:
The strongly correlated electronic system is an exciting and diverse field in condensed matter physics. Electrons are usually localized at their atomic sites in this system having strong electron-electron interaction (Coulomb repulsion). SrCrO\(_4\) is a strong oxidizing agent which is highly corrosive and insoluble in water. It is used in electrochemical processes to control sulfate concentration of solutions. The nanoparticles of SrCrO\(_4\) were manufactured by the sol–gel technique. This system was helpful in gaining the single crystalline and pure monoclinic phase of the materials which was confirmed by the XRD results. SrCrO\(_4\) having space group P2\(_1\)/n. We calcined it at 950°C temperature. Ba and Ca-doped SrCrO\(_4\) were also synthesized by sol-gel method at different concentrations for x= 0.2, 0.4, 0.6 and 0.8. Different characterization techniques such as X-Ray Diffraction, UV-VIS Spectroscopy, PL Spectroscopy and FTIR Spectroscopy were used to analyze SrCrO\(_4\), Ba and Ca-doped SrCrO\(_4\). XRD tells us about the crystal size and dislocation density of samples. The value of 2θ for the XRD patterns is ranging from 5 to 95. Four peaks are observed in the UV-spectra of SrCrO\(_4\) which occur at 350nm, 380nm, 700nm and 750nm. The UV band gap of SrCrO\(_4\) is 3.25 eV. This wide band gap makes these materials suitable for performing photo-catalytic activity under visible light illumination. In PL spectra, two peaks are observed one at 480nm and other at 410nm. At 480nm, the energy of emitted photons is 2.5eV, while at 410nm the emitted photons have energy of 3ev. In the FTIR analysis, the internal modes usually displayed by CrO\(_4\) unit are symmetric stretching bond (\(\nu_1\) (A\(_1\))), symmetric bending mode (\(\nu_2\) (E)), asymmetric stretching mode (\(\nu_3\) (F\(_2\))) and asymmetric bending mode (\(\nu_4\) (F\(_2\))). In future, we will use SrCrO\(_4\) in different applications such as in water splitting, in photo-catalytic activities and evaluation of SrCrO\(_4\) for the formulation of primers used in space applications.

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