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جامعة بنغازي
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Influence of annealing on Si-Ge thin films with high concentration of tellurium and zinc

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Abstract:

The elements are distinguished via the basis of energy band gap. Two configurations $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ multilayers were annealed under vacuum at eutectic temperature of 400 °C to form alloys between the deposited layers. The two samples were studied at different temperatures to improve the physical properties of the thin films. Kinetics of the two configurations $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ were studied in the temperature range 308- 673 °K. The results revealed that the crystallization of the thin film of multilayer for the samples increases with increasing the annealing temperature and increasing the zinc content. The influence of annealing on the structure and stability of the samples has been studied by X- ray diffraction. The optical and electrical measurements were carried out after annealing. The electrical conductivity of the two samples was measured as a function of temperature and annealing time. It was found that the electrical conductivity for $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ increases, and the optical gap decrease due to the crystallization effects that occurs only in Ge matrix and with increasing zinc content at high concentration.

Keywords: thin films, annealing, temperature, crystallization, optical gap, electrical conductivity.

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الريقة ذات التركيز العالي من التيلوريوم والزنك (Si-Ge) تأثير التلدين على أغشية

¹د. إبراهيم صالح عبد الحفيظ.

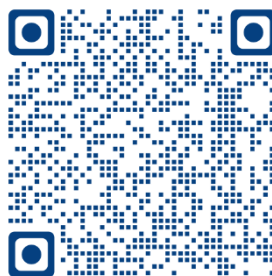
1. أستاذ مساعد بقسم الفيزياء، كلية الآداب والعلوم "الأبيار"، جامعة بنغازي.

المخلص

يتم تمييز العناصر من خلال فجوة نطاق الطاقة الأساسية. تم التلدين بتكوينين $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ و $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ متعدد الطبقات تحت فراغ عند درجة حرارة سهلة الانصهار تبلغ 400 درجة مئوية لتشكيل سبائك بين الطبقات المودعة. تمت دراسة العينتين عند درجات حرارة مختلفة لتحسين الخواص الفيزيائية للأغشية الرقيقة. تمت دراسة حركية التكوينين $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ و $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ في المدى الحراري 308-673 درجة مئوية. أظهرت النتائج أن تبلور الطبقات الرقيقة للعينات يزداد مع زيادة درجة حرارة التلدين وزيادة محتوى الزنك. تمت دراسة تأثير التلدين على بنية وثبات العينات باستخدام حيود الأشعة السينية. تم إجراء القياسات الضوئية والكهربائية بعد التلدين. تم قياس التوصيل الكهربائي للعينتين كدالة لدرجة الحرارة وزمن التلدين. وجد أن الموصلية الكهربائية لـ $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ تزداد، وتقل الفجوة الضوئية بسبب تأثيرات التبلور التي تحدث فقط في مصفوفة Ge ومع زيادة محتوى الزنك عند التركيز العالي.

الكلمات المفتاحية: الأغشية الرقيقة، التلدين، درجة الحرارة، التبلور، الفجوة الضوئية، التوصيل الكهربائي.

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1.Introduction

The modern technological era started with the invention of the transistor and the birth of solid state electronics. From that date, we have been rapidly developed the science and technology based firstly on germanium, then on silicon, and later on multilayers for elements with germanium, and silicon. The necessity to save weight and ratio of element, and to improve the electrical conductivity in the function of the devices, stimulated an intense research activity to obtain the thin films from multilayers. In the same time, the worldwide interest was moved from electronic to more complex optoelectronic applications. Taking into account these big tasks, the class of materials must be enlarged, new materials with new properties must be prepared. The physical properties of thin films materials of many semiconductors and elements has been investigated. [18].

The improvement of devices based on the prepared materials via different high technology fields of science. Nowadays, commercial devices require the development of tailored materials. Approaches to evaluate the physical

properties of their structures produces an inevitable industrial delay in the devices improvement. Moreover, with the trend to continuous curing of the components makes it becomes necessary to deal with the spatial resolution problem. For the advance in such applications, the locally investigation of the change in the electrical conductivity with temperature and elements contents of in new compounds of materials is therefore a key issue.

Materials are commonly affected by a large concentration of point defects, hence the heat treatment are very important. The transition of crystalline state proceeds by nucleation and growth reactions, a systematic study on the kinetics of the crystal phase remains one of the most interesting aspects of physics properties. [7].

A strict control on the ratio of materials and location is necessary, but also because their behavior in the material structure is still rather unclear. Electrical conductivity may also be used to investigate of crystallization for material of thin films and type of elements. [9].

The aim of this work is to study the physical properties of the thin film



s.for.multilayers.and.using.the.results.t
o.investigate.affects.on.the.electrical.co
nductivity.with.annealing.and.high.con
centration.of.materials.contents.

II.Experimental

Two.configurations.of.multila
yers.were.deposited.using.Edwards.3
06.thermal.evaporator.under.vacuum.
of. 6×10^5 .mbar.on.borosilicate.glass.s
ubstrates..The.first.configuration.con
sists.of.silicon(Si),.germanium.(Ge),.
and.tellurium.(Te),.and.the.second.co
nfiguration.consists.of.silicon.(Si),.g
ermanium.(Ge),.and.Zinc.(Zn)..The.
Two.samples.prepared.at. 80°C ...The.
first.sample.in.this.configuration.is.S
 $\text{i}_{10}\text{Ge}_{10}\text{Te}_{80}$.and.the.thicknesses.of.lay
ers.of.this.sample.are.30.nm.of.Si,.30
.nm.of.Ge,.and.240.nm.of.Te..The.se
cond.sample.in.this.configuration.is.
 $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$.and.the.thicknesses.of.l
ayers.of.this.sample.are.30.nm.of.Si,.
30.nm.of.Ge,.and.240.nm.of.Zn..The.
samples.of.the.two.configurations.are
.then.annealed.under.vacuum.at.eute
ctic.temperature.of. 400°C .to.form.al
loys.between.the.deposited.layers..T
he.following.table.shows.the.prepare
d.samples..The.total.film.thickness.m

asured.by.the.Dectak.surface.profile
r.was.in.the.range.of.300nm.

The.crystallinity.of.the.two.samples.
was.measured.by.Xray.diffractomete
r.(.Model:..Labx.XRD6100.).provide
d.by.copper.(Cu).Xray.tube.of.wavel
ength. $\lambda=1.54\text{\AA}$.was.used.for.structue.
analysis.of.the.thin.films..The.interpl
aner.distance.was.determined.using.
Bragg`s.Equation: $2d.\sin\theta=.n.\lambda$
Where.d.is.the.interplaner.distance.a
nd. θ .is.the.angle.of.x-ray.diffraction.
JASCO.V630.UVVIS.Spectrophoto
meter.was.used.for.transmission.and.
absorption..measurements..A.cryosta
t.(.photon.cryostat.220.Ohmmetry.).
was.used.for.electrical.measurements
The.sample.is.maintained.between.t
wo.electrodes.as.a.coplanar.structure.
attached.to.a.dc.voltage.of.60.Volt..T
he.pressure.is.about. 6×10^3 .mbar,.dep
ending.on.the.geometry.of.the.vacuu
m.chamber..The.substrate.holder.is.p
rovided.by.heating.system.to.raise.th
e.temperature.of.the.sample.up.to. 60°C .



III. Results and discussion

1. The X-ray diffraction (XRD)

The structural properties of $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ prepared at 80°C and annealed at 400°C have been investigated by XRD. The phases appeared in the x-ray spectra are GeGe(111), GeGe(2

Table(1): The dspacing of $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ prepared at 80°C after annealing at 400°C for 2h.

Phases	(2 θ). $^\circ$ 2Theta.	d- Spacing.($\text{\AA}$)	$h^2+k^2+l^2$	Orientations	Lattice constant.($\text{\AA}$)	Lattice.Constant.($\text{\AA}$).i n.text.book.[10-11].
Ge(111)	26.889	3.311	3	(111)	5.736	5.74
Ge(211)	38.626	2.328	6	(211)	5.702	5.6575.and.5.74
Ge(220)	44.953	2.014	8	(220)	5.700	5.6575.and.5.74

11),.GeGe(220).for.sample. $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$.and.phases.appeared.in.the.x.ray.spectra.are.GeGe(111),GeGe(211).for.sample. $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$.The.inter.plane r.distances.(dspacing).have.been.calculated.using.the.Bragg's.equation.and.d.are.given.in.table.(1)..

2. Optical Properties

In this section we will discuss the measurements of transmission of $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ thin films prepared at 80°C and the deduced optical energy gap after annealing at 400°C .

2.1..Transmission and absorption measurements.

Figures.(1).show transmission for the $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ thin films after annealing at 400°C . It is clear that for high energies ($\lambda_{\min}$) there is no transmission appeared for samples but for low energies ($\lambda_{\max}$) the transmission is high. For low energies ($\lambda_{\max}$) h

however, there are no appropriate electronic transitions possible so transmission is very high in this range. It is not 100% however, because of absorption. The interpretation of the transmission spectra in figure (1) is not so straightforward. For high energies, where absorption is high according to the transmission chart, it increases as well. This can only be explained by a theoretical treatment of the interaction between light and matter. [12.14].

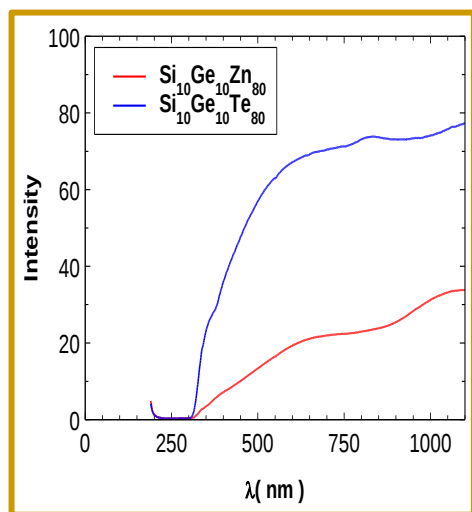


Figure (1): Transmission spectra for untreated the $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ thin films prepared at 80°C and annealed at 400°C for 2 h. It is shown that for high energies ($\lambda_{\min}$) there is no transmission for film Si_{10}

$\text{Ge}_{10}\text{Te}_{80}$, but for film $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ the transmission is about 34%. For high energies ($\lambda_{\min}$), where absorption is high according to the transmission chart. After annealing at 400°C , the absorption increases due to the increasing of crystallization and the high concentration of zinc (Zn) which leads to a decrease of the transmission intensity...

2.2. Optical energy data

The optical band gap is useful material parameter. It allows to compare between the samples $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$ thin films based materials regarding their light absorption properties. [15.17]. The concentration of tellurium and zinc plays an important role for determining electrical conductivity and the optical energy gap for the samples $\text{Si}_{10}\text{Ge}_{10}\text{Te}_{80}$ and $\text{Si}_{10}\text{Ge}_{10}\text{Zn}_{80}$, as conformed by the results obtained in previous works [18]. Figures (2) shows the relation between $(\alpha h\nu)^{1/2}$ and $(h\nu)$ for the two samples. According to Tauc's relation [19], the optical energy gaps were deduced from the plots, and the values are given in tables (2) for the two samples after annealing.

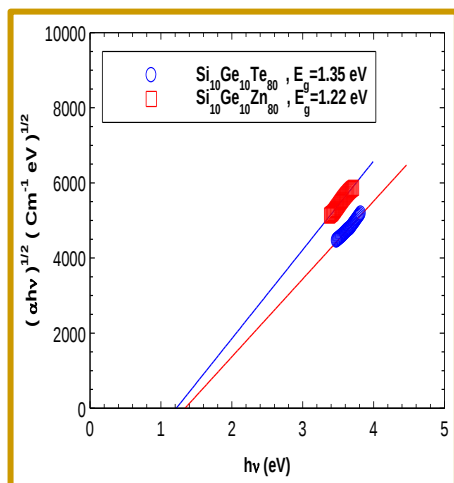


Figure.(2):. $(\alpha hv)^{1/2}$ Vs. $h\nu$ for $Si_{10}Ge_{10}Te_{80}$ and $Si_{10}Ge_{10}Zn_{80}$ thin films prepared at $80^\circ C$ and annealed at $400^\circ C$ for 2 h.

The data shows that the optical energy gap decreases with raising and/or annealing temperature and due to crystallization effects in Ge matrix [20,23], and partially due to high concentration of zinc (.Zn.).

Table.(2):. The data of optical band gap E_g for untreated samples.

The sample	E_g (eV.)
$Si_{10}Ge_{10}Te_{80}$	1.35
$Si_{10}Ge_{10}Zn_{80}$	1.22

3. Electrical properties

3.1. Effects of temperature and (tellurium/zinc) high concentration on the electrical conductivity.

The conductivities as a function of temperature for $Si_{10}Ge_{10}Te_{80}$ and $Si_{10}Ge_{10}Zn_{80}$ thin films prepared at $80^\circ C$ and annealed at $400^\circ C$ for 2 h. Here, the concentration of Te and Zn are 80%, in the temperature range 308.473 K are shown in figures (3). It is seen that the relation between the electrical conductivity and the temperature obey the Arrhenius type equation: $\sigma = \sigma_0 \exp(E_a/kBT)$. (1)

where σ is electrical conductivity, E_a is the activation energy and k_B is the Boltzmann's constant. The high concentration for elements conducting are playing an important role for determining electrical conductivity of multilayers $Si_{10}Ge_{10}Te_{80}$ and $Si_{10}Ge_{10}Zn_{80}$ thin films. The electrical conductivity measured at 308 K and the activation energy calculated from the slopes of the lines for the samples are given in table (3). It is seen that the electrical conductivity increases while activation energy decreases with increasing the content of (.Zn.) more.

than.(.Te.).as.conformed.by.the.previ
ous.works.[18,22,23]..

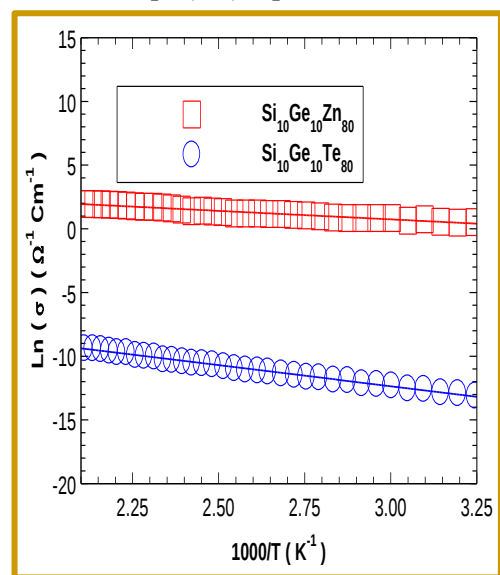


Figure.(3):.Dark.conductivity
.vs..inverse.of.temperature.for.multil
ayers.of.Si₁₀Ge₁₀Te₈₀.and.Si₁₀Ge₁₀Zn₈₀.thin.films.

Table.(3):.Conductivity.meas
ured.at.308.K.and.the.activation.ener
gy.for.untreated.multilayers.of.Si₁₀G
e₁₀Te₈₀.and.Si₁₀Ge₁₀Zn₈₀.thin.films.

The.sample	concentration.(8 0%)	$\sigma(\Omega^{-1}.cm^{-1}).$	$E_a.(eV.).$
Si ₁₀ Ge ₁₀ Te ₈₀	Te	26×10^{-3}	0.29
Si ₁₀ Ge ₁₀ Zn ₈₀	Zn	31×10^{-2}	0.12

3.2..Kinetics.and.thermal.stability

The.electrical.conductivity.of
.multilayers.of.Si₁₀Ge₁₀Te₈₀.and.Si₁₀

Ge₁₀Zn₈₀.thin.films.as.a.function.of.a
nnealing.times.were.recorded.at.diffe
rent.temperatures.473,573.and.673.K
.is.given..figure.(4).and.(5),.respective
ly..All.samples.show.the.same.gener
al.behavior.where.the.electrical.con
ductivity.increases.with..increasing.t
he.annealing.time..at.constant.anneal
ing..temperature.and.becomes.consta
nt.at.high.annealing.time..This.behav
ior.is.attributed.partially.to.crystalliz
ation.occuring.in.the.multilayers,.th
at.crystallization.effects.occur..only.i
n.Ge.matrix.with.annealing..temperat
ure..and..annealing.time..as..confirm
ed.by.Xray.data,..and.also.depending
.on.the.incorporation.of.zinc.or.tellur
ium.at.the.network.

It.is.known.that.the.conductiv
ity.of.crystalline.material.is.higher.th
an.that.of.amorphous.one.since.the.or
dered.systems.exhibit.lower.activatio
n.energy.than.the.amorphous.one.[24
].The.study.confirms.that.electrical.c
onductivity.increases.with.increases.
at.high.concentration.of.the.element.
doping..This.is.clearly.for.the.sample
.doped.with.zinc.at.high.concentratio
n,.more.than.for.the.sample.doped.wi
th.tellurium..Thus..the.electrical.cond

activity..measurements..as.a.function
..of..annealing..time.at.constant..tem
perature..are.used.to..study.the.isothe
rmal.crystallization..kinetics.using.Jo
hnson.Mehl.Avermi`s.(JMA).equatio
n..

Thus.the.electrical.conductivi
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ealing.time.at.constant.temperature.a
re.used.to.study.the.isothermal.crysta
llization.kinetics.using.Johnson-
Mehl-.Avermi`s.(JMA).equation.in.t
he.form[25]:.

$$\chi = 1 - \exp[-kt^n] \quad (2)$$

Where. χ .is.the.volume.fraction
n.of.the.crystalline.phases.transforme
d.from.the.amorphous.state.at.time.t.,
n.refers.to.the.order.of.reaction.and.k
.is.the.effective.overall.reaction.rate.,
which.actually.reflects.the.rate.of.cry
stallization.[26,.27].

The.electrical.conductivity.as
.a.function.of.annealing.time.the.volu
me.fraction. χ .is:.

$$\chi = (\ln \sigma_a - \ln \sigma_t) / (\ln \sigma_a - \ln \sigma_c) \quad (3)$$

Where. $\ln \sigma_a$.is.logarithm.of.th
e.electrical.conductivity.at.zero.time.
(activation.electrical.conductivity),. $\ln$
 σ_t .logarithm.of.the.electrical.condu
ctivity.at.any.time.t.and. $\ln \sigma_c$.is.logar

ithm.of.the.electrical.conductivity.at.
the.end.of.saturation.(full.crystallizat
ion).

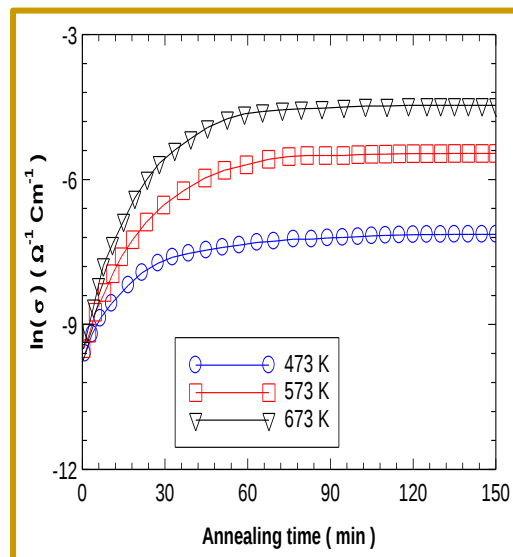


Figure.(4):.logarithm.of.the.el
ectrical.conductivity.versus.the.anne
aling.time.at.constant.temperatures.f
or.Si₁₀Ge₁₀Te₈₀.multilayers.

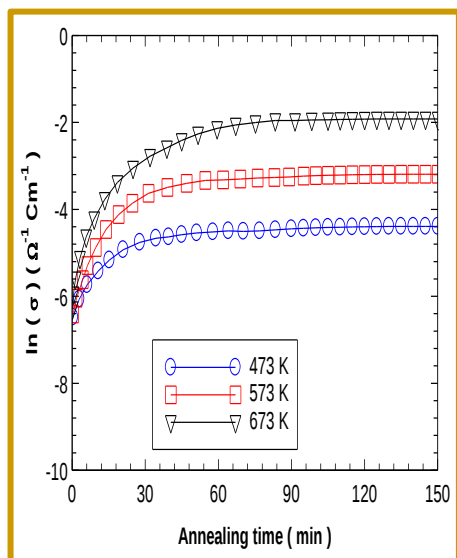


Figure.(5):.logarithm.of.the.el
ectrical.conductivity.versus.the.anne
aling.time.at.constant.temperatures.f
or.Si₁₀Ge₁₀Zn₈₀.multilayers.

IV.Conclusions

The.samples.show.the.same.g
eneral.behavior.where.the.electrical.c
onductivity.increases.with..increasing.
the.annealing.time.at.constant.anneali
ng..temperature..and..becomes.consta
nt.at.high.annealing.time..This.behavi
or.is.attributed.partially.to.crystallizati
on.occuring.in.the.multilayers,.especi
ally.for.germanium.atoms.depending.
on.the.annealing.temperature.and.ann
ealing.time.as.confirmed.by.Xray.dat.,
and.due.to.the.incorporation.of.zinc.or

.tellurium.to.the.network..The.elemen
ts.of.dopes.at.high.concentration.plays
.an.important.role.for.determining.the.
electrical.conductivity.and.optical.ga.i
n.this.study.the.conductivity.are.(p_{type}
e).similarly.for.holes..The.reason.in.c
hanging.optical.gap,.this.may.be.due.t
o.shifting.the.Fermi.level,.where.incre
asing.shifting.the.Fermi.level.to.wards
.the.valence.band.upon.zinc.or.telluriu
m.doping.in.the.thin.films..

It.was.found.the.electrical.co
nductivity.increasing..and.the.optical.
gap.decreasing.with.the.crystallization
..effects.occur.only.in..Ge.matrix.and.
with..increasing.zinc.at.high.concentr
ation..Where.the.optical.gap.change.fr
om.1.35eV.to1.22eV.at.high.concentr
ation.for.tellurium.and.zinc.respective
ly..It.is.clear.that.the.change.of.the.ele
ctrical.conductivity.with.annealing.ti
me.at.different.isothers.is.one.of.the
.sensitive.physical.properties.to.reflec
t.the.change.and.growth.of.the.phase.t
ransformation.process.of.any.material

V..Acknowledgements

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Mohamed.Nawwar,.Dr..of.solid.state
.physics,.Physics.Department,.Facult
y.of.Science,.Menoufia.University,.E



gypt.for.his.kind.an.immensely.help.i
n.achieving.my.goals..This.work.is.s
ponsored.by.the.Libyan.ministry.of.h
igher.education.and.Benghazi.Univer
sity..

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