9h. Color Centers and Dislocations

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9h-1. General Properties of Color Centers. Color centers are imperfections in transparent solids that give rise to optical absorption. Most of these centers are associated with crystalline defects, but centers arising from the incorporation of chemical impurities are frequently also considered to be color centers. Work on this subject has progressed farthest in the alkali halides; and only these materials will be discussed here. Related centers appear in most transparent solids, but their atomic identification is uncertain in many cases.

The conditions under which each center appears will be discussed in more detail below for the various centers. The most common treatments (ref. 1) are exposure to ionizing radiation such as X rays, heating in the alkali metal vapor which leads to centers with trapped electrons, and heating in halogen vapor which leads to centers with trapped holes.

It is frequently useful to relate *the number of centers* to the strength of the absorption band produced. If the absorption band is gaussian in shape, then an approximate relation is (ref. 2)

$$N_0 f = 8.7 \times 10^{16} \frac{n}{(n^2 + 2)^2} \alpha_m W \tag{9h-1}$$

where N_0 is the concentration of centers per cubic centimeter, f is the oscillator strength, n is the index of refraction of the material at the wavelength of the absorption band, α_m is the absorption coefficient at the maximum of the band in reciprocal centimeters, and W is the width of the absorption band in electron volts at an absorption coefficient one-half that of the maximum. If it is possible to measure the concentration N_0 by some method such as chemical analysis or magnetic susceptibility, then the oscillator strength can be obtained. Knowledge of this factor for a particular center allows the determination of the density of that center from optical measurements alone and also gives a measure of the degree to which the optical transition is an allowed one. If the curve is Lorentzian in shape, then the constant in Eq. (9h-1) is 12.9. This form of the equation, often called Smakula's equation, is used in much of the older work, but the Gauss curve is a better (refs. 3 and 4), but not perfect, fit to the observed bands. Oscillator strengths given here will be in terms of Eq. (9h-1); they can be converted to Smakula's equation by multiplying by 1.48.

9h-2. F-center and Other Trapped Electron Centers. The most widely investigated color center is the *F-center* now known *to consist of an electron trapped at a negative ion vacancy*. If an alkali halide crystal is heated in the vapor of the alkali metal for several hours and then quenched to room temperature, the F-band appears. To the short-wavelength side of the F-band there also appear several weak absorption bands which have been designated K-, L_1-, L_2-, and L_3-bands. It is believed that these are more highly excited states of the F-center and show a dependence of wavelength on lattice constant which is similar to that of the F-band (ref. 5).

If the F-band is bleached with light at low temperatures (−150°C for KBr, for example) a new broad band grows to the long-wavelength side of the F-band. This absorption is due to the *F'-center* and *arises from an F-center that has captured an additional electron* (ref. 6).

Irradiation with light in the F-band at room temperature causes the F-band to decrease and produces the *M-band* which arises from *a pair of F-centers*, and then the R_1- and R_2-bands arising from *a cluster of three F-centers*.

The peak position of the absorption of these centers at room temperature is given in Table 9h-1. Wavelengths are given throughout this article in millimicrons (mμ).

TABLE 9h-1. WAVELENGTH OF ABSORPTION OF ELECTRON TRAP CENTERS
(In millimicrons)

	L_3 −180°C	L_2 −180°C	L_1 −180°C	K −180°C	F 20°C	R_1 20°C	R_2 20°C	M 20°C	Width at half maximum of F-band, eV 20°C
LiF......	...	...	...	...	250	313	380	444	0.82
LiCl.....	...	...	...	...	385	...	580	650	0.62
NaF.....	...	...	...	...	341	...	415	505	0.62
NaCl....	...	...	...	...	458	545	596	725	0.47
NaBr....	...	...	...	...	540	...	...	...	0.52
NaI.....	...	...	...	...	588				
KF......	...	...	...	...	455	...	570	...	0.41
KCl.....	251	288	344	457	556	658	727	825	0.35
KBr.....	276	316	374	525	625	735	790	892	0.345
KI......	326	382	447	585	689	...	...	...	0.345
RbCl....	279	335	402	523	609	...	...	...	0.31
RbBr....	300	362	435	593	694	805	859	957	0.28
RbI.....	338	413	506	640	756	...	...	...	0.35
CsCl....	...	...	...	...	605				
CsBr....	...	...	...	...	680				

The values are somewhat approximate, since different workers report results varying by as much as 20 mμ. Also given in Table 9h-1 are the widths at half maximum of the F-band at room temperature.

It has been noted that the wavelengths of the absorption bands vary with the distance a between nearest neighbors of the alkali halides. Equations (sometimes called Ivey relations) governing these bands are as follows (ref. 7):

F-center: $\lambda_{abs} = 703a^{1.84}$

R_1-center $\lambda_{abs} = 816a^{1.84}$

R_2-center: $\lambda_{abs} = 884a^{1.84}$

M-center: $\lambda_{abs} = 1{,}400a^{1.56}$

Both λ and a are in angstroms.

The variation of the maximum of the F-center absorption band as a function of temperature is shown in Fig. 9h-1 (ref. 16). The width W at half maximum of the F-band absorption also varies with temperature and fits an equation of the form

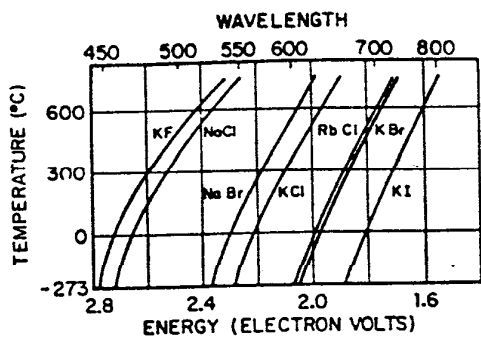$$W = W_0 \coth\left(\frac{h\nu}{2kT}\right)^{\frac{1}{2}} \qquad (9h\text{-}2)$$

FIG. 9h-1. Effect of temperature on the position of the F-band maximum.

where W_0 is the width at absolute zero, h is Planck's constant, ν is a frequency related to the lattice vibrations of the solid, k is Boltzmann's constant, and T is the absolute temperature. At low temperatures (less than about 25° absolute) W is a constant; at high temperatures (above room temperature) W increases with the square root of the temperature. Table 9h-2 lists the values of W_0 and ν which give the best fit to experiment (ref. 17). W and W_0 are given in units of electron volts.

Direct measurements of oscillator strengths have been made for some of the F-centers using chemical techniques, electron-spin resonance measurements, and measurements

TABLE 9h-2. HALF WIDTH OF F-BAND AS A FUNCTION OF TEMPERATURE
[Constants for Eq. (9h-2)]

	W_0, eV	ν, Hz
LiF............	0.43	4.1×10^{12}
NaCl..........	0.29	4.4×10^{12}
KCl...........	0.18	2.6×10^{12}
KBr...........	0.20	2.6×10^{12}
KI............	0.18	3.6×10^{12}

of paramagnetic susceptibility. The agreement among these various methods is relatively poor. Oscillator strengths of the following values have been reported:

F-center in NaCl: 0.5[8], 0.58[9], 0.5[10], 0.57[11]
F-center in KCl: 0.55[8], 0.54[12], 0.57[5], 0.44[10], 0.78[13], 0.61[11]
F-center in KBr: 0.47[10], 0.57[11]
F-center in KI: 0.31[10]

From measurements on the growth of the M-band as the F-band is bleached by light, values of 0.2 (refs. 14 and 15) have been obtained for the oscillator strength of the M-band in KCl.

The effect of pressure on the position of the F-center maximum has been measured (ref. 18) in the range up to 50,000 atm. Data obtained at room temperature for the F-band in NaCl, KCl, and CsBr are shown in Fig. 9h-2. In the case of KCl the sharp
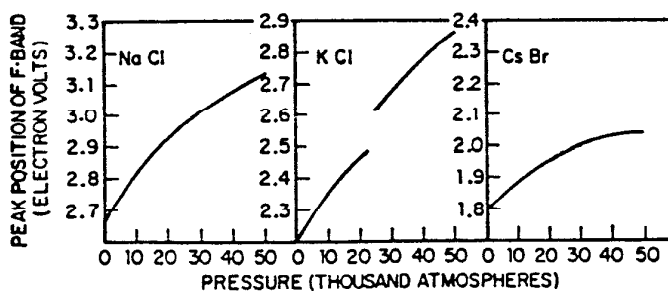

FIG. 9h-2. Effect of pressure on the position of the F-band maximum.

break in the curve occurs as the lattice changes from the normal NaCl type to that of CsCl at high pressures.

When color centers are excited, the stored energy may be released as luminescence. If only F-centers are present and measurements are made at liquid-nitrogen temperatures or below, F-center luminescence is observed (ref. 19). When M-centers are present, then excitation in both the F-band and M-band gives rise to M-center emission. Finally, when the R-centers arise, only emission characteristic of these centers can be observed. In general only the M-center emission can be seen on exciting at room temperature.

From measurements of the polarization of luminescence emission as a function of the polarization of the exciting light, the symmetry of the centers can be obtained. The F-center is found to be isotropic, but the M- and R-centers are asymmetrical and have their major axis along the $\langle 110 \rangle$ directions. Table 9h-3 gives the luminescence

TABLE 9h-3. LUMINESCENCE OF ELECTRON TRAP CENTERS AT 77 K

	F-center		M-center			R-center		
	Peak position, mμ	Half width, eV	Peak position, mμ	Half width, eV	P	Peak position, mμ	Half width, eV	P
LiF......	.....	....	670	....	0.60			
NaCl.....	1,200	0.31	1,070	0.20	0.60	1,180	0.19	0.20
KCl......	1,010	0.20	1,080	0.21	0.60	1,240	0.15	0.20
KBr......	1,320	0.22						
KI.......	1,470	0.16						
RbCl.....	1,120	0.23						

peak positions and half widths for various centers and gives also the polarization P of the luminescence of the M- and R-centers (ref. 20). This polarization is measured with the exciting light, polarizer, sample, analyzer, and detector in line. If the polarizer is set parallel to a $\langle 110 \rangle$ direction, then the luminescence measured with the analyzer parallel to the polarizer is $I_\parallel$ and with the analyzer crossed is $I_\perp$. The polarization P is defined as

$$P = \frac{I_\parallel - I_\perp}{I_\parallel + I_\perp}$$

For simple dipoles along the $\langle 110 \rangle$ direction, the value of P should be 0.66.

Electron-spin resonance has been observed for the F-center in many of the alkali halides. Table 9h-4 gives the g-values of the resonance (compared with a value of 2.023 for the free electron) and the width ΔH of the resonance absorption. This value is expressed in gauss for measurements in which the magnetic field is approximately 3,000 gauss and the frequency is approximately 9,000 MHz.

9h-3. Hole Trap Centers. A group of centers exists in the alkali halides characterized by having trapped a hole. Transfer of an electron from an electron trap center to one of these hole trap centers destroys them both. These centers do not follow an Ivey relation. Although some of them are formed by additive coloration at high temperature, the best-understood ones are formed at low temperatures by X-raying and show very detailed paramagnetic spectra. The peak position of the absorption bands for some of these centers is given in Table 9h-5.

The H-center is formed by X-raying at liquid-helium temperatures. From a study of its detailed paramagnetic resonance spectrum (ref. 25), the *H-center is found to be three halide ions and a halogen atom squeezed into the position normally occupied by three halide ions along a face diagonal*. The center bleaches thermally at 56 K in KCl.

X-raying at 77 K produces the V_1-center, an H-center near an impurity. Also at 77 K a weak absorption band, called the X_2^--band, is formed. The number of these centers is much larger if small amounts of Ag, Tl, or Pb are in the crystals (ref. 26). From a study of the paramagnetic resonance spectra (ref. 27) it is concluded that the X_2^--center consists of a hole trapped between two halogen ions which have been displaced toward each other slightly from their equilibrium positions.

TABLE 9h-4. ELECTRON-SPIN RESONANCE OF THE F-CENTER

Material	g-value	ΔH	Ref.
LiF	2.003	120	21
NaF	2.002	50	21
NaCl	1.987	180	22
KCl	1.995	61	22
KBr	1.980	162	22
KI	1.971	200	23
RbCl	2	400	24
RbBr	2	380	24
RbI	2	640	24

TABLE 9h-5. WAVELENGTH OF ABSORPTION OF HOLE TRAP CENTERS
(In millimicrons)

	H 4 K	V_1 77 K	X_2^- 77 K	V_2 300 K	V_3 300 K
LiF	...	...	348		
NaCl	330	345	...	223	210
KCl	335	356	365	230	212
KBr	380	410	385	265	231
KI	...	...	404		

The V_2- and V_3-centers are formed in alkali halides by heating them in halogen vapor and quenching to room temperature.

9h-4. Perturbed Lattice Transitions. Two bands, the α- and β-bands, have been found in alkali halides near the edge of the fundamental absorption band upon X-raying at liquid-nitrogen temperatures (ref. 28) (Table 9h-6). The strength of the α-band is correlated with the presence of negative ion vacancies and the β-band with F-centers. It is believed that both bands arise from transitions similar to those in the fundamental band of pure crystals but modified by the proximity of the various imperfections. The oscillator strength of the β-band is approximately unity, and that of the α-band somewhat less.

9h-5. Colloid Centers. Colloid centers are formed in crystals that have been colored by heating in alkali vapor and are then held at temperatures between 300 and 500°C. An absorption band to the long-wavelength side of the F-band appears. As the temperature increases over this range, the F-band intensity increases, the colloid

band decreases, and its peak position shifts to longer wavelength. It is believed that these bands are due to colloid metal particles of from 10 to 50 Å in diameter.

9h-6. Impurity Absorption Bands. Alkali halide crystals containing hydrogen show an absorption band known as the U-band. The *U-center consists of a hydride ion substituting for a normal halide ion.* Irradiation with light in the U-band produces the F-center and a new center, the U_2-center, *due to interstitial hydrogen atoms. Interstitial hydrogen ions give rise to the U_1-center* (ref. 29). The absorption peak positions of the U-bands are given in Table 9h-7. For KCl the U_2-band occurs at 236 mμ and the U_1-band is a broad band near 275 mμ.

TABLE 9h-6. ABSORPTION OF α- AND β-BANDS
(Absorption peaks in millimicrons)

	α	β
NaF	131	127
NaCl	173	168
NaBr	199	
KCl	178	170
KBr	201	192
KI	238	226
RbBr	205	196
RbI	240	229

TABLE 9h-7. ABSORPTION BANDS FROM U-CENTERS, HYDROXIDE CENTERS, Z_1- AND Z_2-CENTERS
(In millimicrons)

	U-band	OH⁻-band	Z_1 band	Z_2 band
NaCl	192	185	505	512
NaBr	210			
KCl	214	204	590	635
KBr	228	214		
KI	244			
RbCl	229			
RbBr	242			

Incorporation of OH⁻ in alkali halides gives rise to the *hydroxide center* absorption bands shown in Table 9h-7, which follow an Ivey relation (refs. 30 and 31). The presence of these bands influences the amount of F-center coloration by X rays at room temperature and the formation of colloids (ref. 32).

Z-centers are formed from additively colored crystals that are doped with divalent impurities such as strontium, barium, or calcium (ref. 33). The Z_1-center can be formed by irradiating an additively colored crystal in the F-band at room temperature. The Z_2-center is formed by heating a crystal containing F- and Z_1-centers to approximately 100°C. Positions of these absorption bands are given in Table 9h-7. The Z_2-center in KCl is luminescent and emits at 1,140 mμ; the emission does not appear to be polarized (ref. 34).

The addition of heavy metal ions to the alkali halides produces absorption and emission bands largely characteristic of the ions. Table 9h-8 shows the optical properties of the centers due to incorporation of Tl, Pb, Ag, and Cu.

TABLE 9h-8. ABSORPTION AND EMISSION BANDS DUE TO Tl, Pb, Ag, AND Cu
(In millimicrons)

	Tl abs.	Tl emis.	Pb abs.	Pb emis.	Ag abs.	Ag emis.	Cu abs.	Cu emis.
NaCl	199 254	288 ...	193 274	318 384 453	210	249	255	358
NaBr	216 267	295 308	220 ...	... ...	219 ...	263 ...	259 ...	365 438
NaI	234 293							
KCl	195 247	250 305 475	196 273	346	215	272	265	396
KBr	210 261	318 350	223 302	...	...	...	265	393
KI	236 287	415	265					
RbCl	195 245	315	198					
RbBr	212 259							
RbI	240 286							
CsCl	106 248							
CsBr	214 263							
CsI	241 269 299							

References for Secs. 9h-1 through 9h-6

There are a series of survey articles and books on color centers each of which gives an excellent summary of the field at the time of its publication. They are listed below in chronological order.

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9h-7. Dislocations. There are two important simple types of dislocations in crystals: the *edge dislocation* and the *screw dislocation*. Figure 9h-3 illustrates an edge dislocation. In this case one portion of the crystal has partially slipped with respect to the other. The plane along which the slip has occurred is plane *ABCD*. If the slip has been one atom distance, then all the atoms are once again in order except for those along the line *AD*. This line, along which the crystal is badly distorted, is
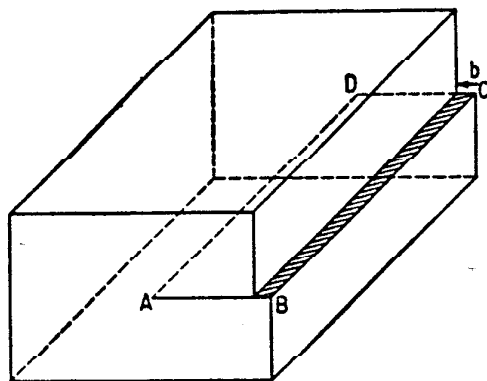

Fig. 9h-3. An edge dislocation. (*After W. T. Read, "Dislocations in Crystals," McGraw-Hill Book Company, New York, 1953.*)
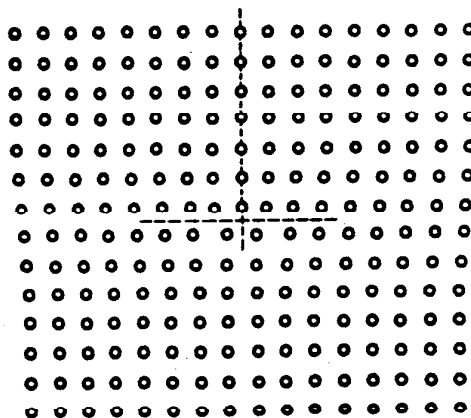
Fig. 9h-4. View of the surface of a crystal with an edge dislocation. (*Reprinted with permission from W. Shockley, J. H. Hollomon, R. Maurer, and F. Seitz: "Imperfections in Nearly Perfect Crystals," John Wiley & Sons, Inc., New York, 1952.*)

the dislocation. The direction of displacement of the atoms due to the formation of the dislocation is *b*, the *Burgers vector*, and for an edge dislocation *b* is always perpendicular to the dislocation line. Figure 9h-4 shows an end view of the crystal where the dislocation line comes through to the surface at *A*. The *slip plane* is represented by the horizontal line. It is seen that the edge dislocation can also be thought of as consisting of the partial introduction of an extra vertical plane of atoms. The end of this plane is the dislocation line.
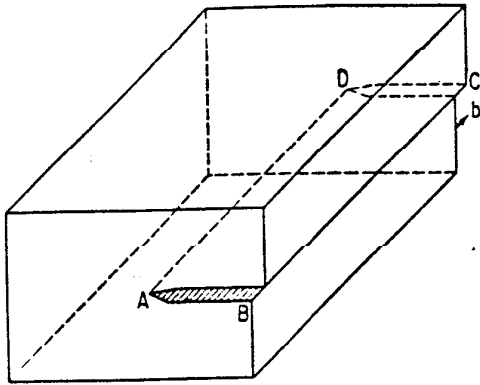
FIG. 9h-5. A screw dislocation. (*After W. T. Read, "Dislocations in Crystals,"* McGraw-Hill Book Company, New York, 1953.)

A screw dislocation is illustrated in Fig. 9h-5, and the surface of the crystal through which it appears is shown in Fig. 9h-6. Here the Burgers vector b is parallel to the dislocation line.

More general forms of dislocation lines are possible. A ring, for instance, consists of portions which are pure edge and pure screw dislocations connected by segments which have both edge and screw character.

The Burgers vector can be obtained in general by making a circuit around a dislocation. Starting in an undistorted part of the crystal we might proceed by counting up l atoms, then left r atoms, then down l atoms, and right r atoms. If this bounded surface does not contain a dislocation, one arrives at the starting point. If a dislocation line does pass through the surface, the circuit will not close on the origin. The vector necessary to close the circuit is the Burgers vector.

The large amount of slip observed along single planes necessitates that there be a source for the creation of many dislocations within a strained crystal. One such model is the *Frank-Read source* illustrated in Fig. 9h-7. The line is a dislocation pinned at both ends by the presence of an impurity atom, for instance. Under stress
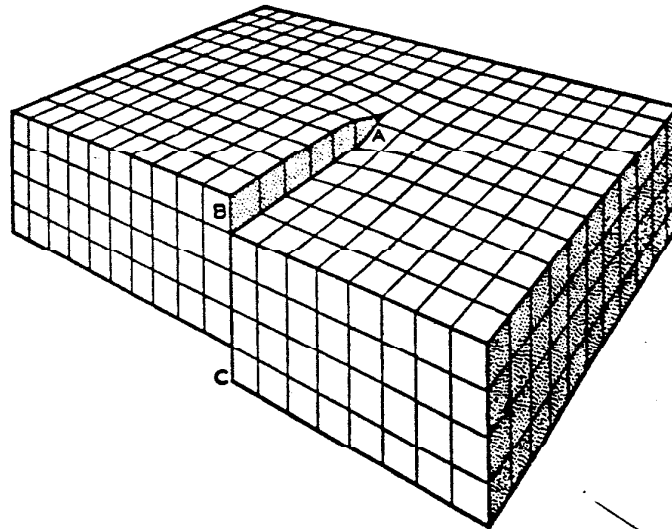

FIG. 9h-6. View of the surface of a crystal with a screw dislocation. (*Reprinted with permission from W. T. Read, "Dislocations in Crystals,"* McGraw-Hill Book Company, New York, 1953.)

the line bows out and finally curls back on itself to touch. One loop continues on; the other returns to the original configuration from which the process may be repeated.

A *jog* in a dislocation is a sidewise step, usually of one atomic distance, of a dislocation line.

Dislocations have been made visible in silver halides and alkali halides by appropriate treatment. It is found that they generally appear to form a hexagonal network. A convenient way to determine the number of dislocations in a crystal is to
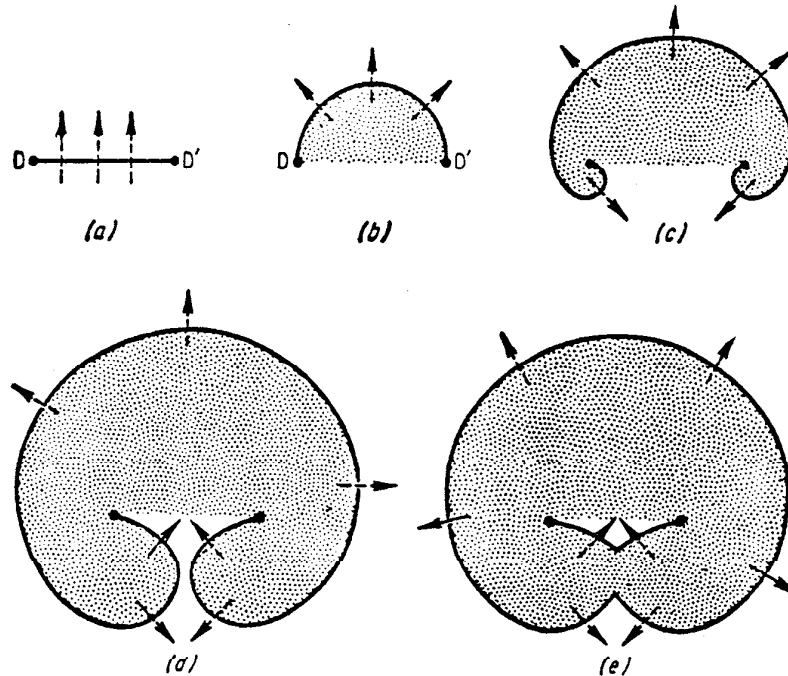
FIG. 9h-7. A Frank-Read source for dislocations. Steps *a* to *c* show the formation of a dislocation loop under a shear stress. (*Reprinted with permission from W. T. Read, "Dislocations in Crystals," McGraw-Hill Book Company, New York. 1953.*)

etch the surface and count the ends of dislocation lines which appear as etch pits The density of dislocation lines is given as those passing through a square centimeter. Normal crystals have approximately 10^6 to 10^9 dislocations per square centimeter, and this may be reduced in very carefully prepared crystals to a few hundred or less.

Etching also allows the motion of a single dislocation to be followed as stress is applied and thus permits studies of the mobility of dislocations.

Dislocations have made it possible to understand the growth of crystals under conditions of very low supersaturation. If a screw dislocation intersects the surface, as in Fig. 9h-6, then the atoms can easily continue to build on the spiral. When examined carefully with the microscope, a great many crystals show this spiral growth pattern.

Another problem solved by dislocations is that of the plastic flow of metals which occurs at stresses less by a factor of 10^4 than those calculated for a pure crystal. The relatively easy motion of dislocation lines has now been used to explain a large variety of mechanical properties.

In recent years it has been possible to obtain dislocation-free crystals. These are usually in the form of thin small needles called *whiskers*. It can be shown experimentally that these whiskers have the mechanical properties expected of the pure materials. Whiskers of many materials have been prepared, including Fe, Cu, CdS, *p*-toluidine, and the potassium halides. While most of the whiskers are straight, a commonly observed defect is a sharp bend, or *kink*, in the crystal.

References for Sec. 9h-7

A number of detailed expositions of dislocations are available.

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