



Mechanism of Adhesive –Free Bonding (AFB[®])

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1.1. Background

AFB[®] composites can consist of similar crystals such as doped and undoped YAG or dissimilar composites. The study of dissimilar AFB[®] composites, e.g. of crystals of different chemical composition and coefficients of thermal expansion (CTE) results in more information to interpret, ultimately leading to the explanation of bonding phenomena for both similar and dissimilar composites.

Dissimilar AFB[®] crystal composites remain essentially stress-free after being heat treated to elevated temperatures that are required by the process. The fact that the composites acquire no net differential thermal expansion-induced stress at the interfaces after being heat-treated is an exception rather than a rule for bonded dissimilar composites. We hypothesize that the composites must relieve the stress by sliding against each other at the interface. We have found experimentally that the thermal shock resistance of stress-free dissimilar composites is the same as that of un-bonded counterparts if not greater. The attraction exerted at the interface of composites must be strong enough to stay intact yet allowing slide upon stress. We attribute this characteristic to the nature of the long-range intermolecular bonds formed at the interfaces.

The long-range intermolecular forces include various modes of Van der Waals forces that exist for all materials in close proximity. The magnitude of the force between two solid bodies is a function of the separation distance and of a material constant denoted as Hamaker constant.

We have produced stress free dissimilar AFB[®] crystal composites of all binary permutations of the following crystals: YAG, sapphire, spinel, and GGG. We have also obtained favorable results with dissimilar composites with CVD diamond as one component.

The occurrence of intermolecular forces and the conditions that lead to formation of direct Van der Waals bonds is investigated comprehensively in the following. We determine general criteria for finding compatibilities between dissimilar materials that will allow them to form strong composites. We then set up experiments that allow *in situ* observation of stress relieving mechanism during thermal treatment experiment as means of validating the theory.

1.2 Occurrence of intermolecular forces and formation of Van der Waals bonds

Most of the intermolecular forces enacted between two dielectric bodies which we encounter in optical components are largely Van der Waals forces. Van der Waals forces consist of three types; they are fixed dipole/ fixed dipole interactions, fixed dipole/ induced dipole interaction, and induced dipole/ induced dipole interactions, respectively [1,2,3].

The Hamaker model describes the interactive potential between two semi-infinite bodies via two flat surfaces of a small but finite separation distance by the following equation.

$$\Phi(x) = -A_{1/3/2} * f(x) \quad \text{eq. 1}$$

Where:

$f(x) = 1/(12 * \pi * x^2)$ for two semi-infinite bodies (1 and 2) being separated by a medium 3.

$A_{1/3/2}^*$ = the Hamaker coefficient of the system 1/3/2.

Where the system 1/3/2 is shown schematically in **Figure 1**

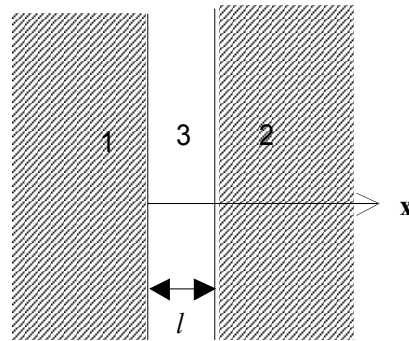


Figure 1. This schematic drawing shows the contacting interface system of 1/3/2 where 1 and 2 are semi-infinite contacting solid bodies and 3 is the material medium-filled interface gap of width of $x = l$. Medium 3 can be e.g. air or vacuum.

The Lifshitz theory [4,5] treats the problem fundamentally by the following premises.

- 1) Randomly fluctuating electromagnetic waves exist everywhere anytime and can be quantized and characterized statistically by their correlation function.
- 2) Van der Waals forces arise due to the interaction between the matter and the random field that follows the Maxwell equations,
- 3) The Van der Waals forces become prominent when the separation distance between two adjacent solid bodies becomes much smaller than any wavelength of the random field, and
- 4) The Van der Waals attraction between the two adjacent solid bodies is the manifestation of Maxwell stress within the system.

The derived expression of the interactive potential by this theory can correlate to the Hamaker model shown in eq.1. The Hamaker constant can then be calculated according to the equation shown below. It requires only the absorption spectrum of the materials as the input.

$$-A_{1/3/2} \sim (3/2)kT \sum_{n=0}^{\infty} \Delta_{13}(i\xi_n) \Delta_{23}(i\xi_n)(1+r_n)\exp(-r_n) \quad \text{eq. 2}$$

where: $r_n = (2/\epsilon_3(i\xi_n))^{1/2} \mu_3^{1/2}/c \xi_n$ the dimensionless distance in x direction

$\xi_n = (2\pi kT/h) * n$ where $n = 1, 2, \dots$ the discretized frequency

$$\Delta_{13}(i\xi_n) = \epsilon_1(i\xi_n) - \epsilon_3(i\xi_n) / (\epsilon_1(i\xi_n) + \epsilon_3(i\xi_n))$$

$$\Delta_{23}(i\xi_n) = \epsilon_2(i\xi_n) - \epsilon_3(i\xi_n) / (\epsilon_2(i\xi_n) + \epsilon_3(i\xi_n))$$

$$\text{where } \epsilon_j(i\xi_n) = 1 + 2/\pi \int_0^\infty (v * \epsilon_j''(v) / (v^2 + \xi_n^2)) dv$$

where: $\epsilon_j''(v)$ is the imaginary part of the complex dielectric function of j at frequency v.

The values of $\epsilon_j(i\xi_n)$ can be deduced from the absorption spectrum of the material j with relative accuracy. With the absorption spectrum of the material known in mainly in the UV to IR spectra range, one can estimate the values of $\epsilon_j(i\xi_n)$ by the following equation.

$$\epsilon_j(i\xi_n) = 1 + C_{IR} / [1 + (\xi/w_{IR})^2] + C_{UV} / [1 + (\xi/w_{UV})^2] \quad \text{eq. 3}$$

where C_{IR} and C_{UV} are the absorption strength in the IR and UV ranges, respectively. ω_{IR} and ω_{UV} are the center frequencies of the absorption bands in IR and UV, respectively (**Figure 2**).

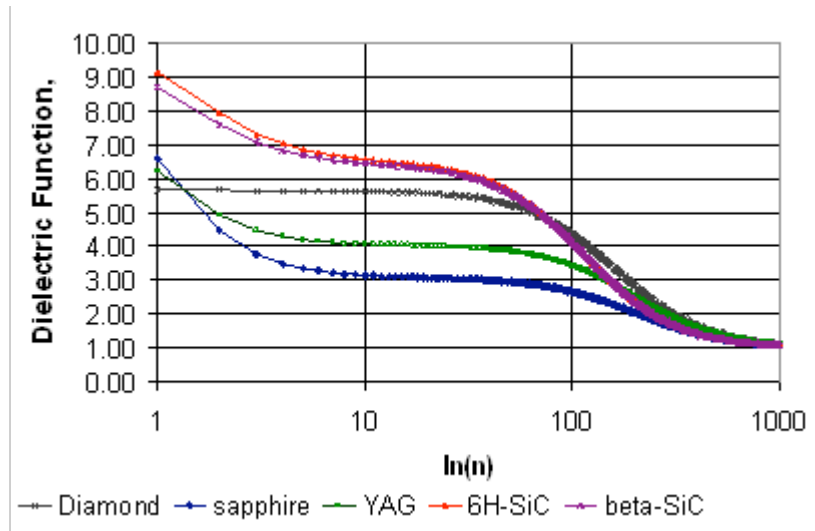


Figure 2. Dielectric function of j for diamond, sapphire, YAG and SiC

An even simpler yet sufficiently accurate model for calculating $\epsilon_j(i\xi_n)$ for two similar materials separated by air is shown as the equation below.

$$A_{1/3/1} = (3/2)kT * \{[(\epsilon_1(0)-1) / (\epsilon_1(0)+1)] + (3h\omega_{UV}/(32*\pi*\sqrt{2})) * \{(n_1^2-1)2/[(n_1^2+1)^{3/2}]\} \} \quad \text{eq. 4}$$



Some of the Hamaker coefficients relevant to solid-state laser hosts have been published over the years. Some of the constants pertaining to this study are listed as follows:

$A_{\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3} = 168.7 \text{ zJ}$, $A_{\text{spinel}/\text{spinel}} = 126 \text{ zJ}$, $A_{\text{SiO}_2/\text{SiO}_2} = 66.6 \text{ zJ}$, $A_{\text{diamond}/\text{diamond}} = 296 \text{ zJ}$ where $1 \text{ zJ} = 10^{-21} \text{ J}$. The Hamaker constant of YAG, $A_{\text{YAG}/\text{YAG}}$, while important as lasing host crystal, is apparently absent in the literature. Given the Hamaker constants of similar crystal composites, one can also calculate the Hamaker coefficients for dissimilar composites using the following equations:

$$A_{1/3/2} = (A_{1/3/1} * A_{2/3/2})^{1/2} \quad \text{eq.4.1}$$

For example, we have calculated the Hamaker coefficient of the diamond/ Al_2O_3 system, $A_{\text{diamond}/\text{Al}_2\text{O}_3}$ to be 223.5 zJ. A Hamaker constant of $A=227 \text{ Jz}$ has been experimentally derived for Yb:YAG/air/Yb:YAG, with the separation distance between Yb:YAG/air/Yb:YAG identical to that of sapphire/vacuum/sapphire as the only assumption. Hamaker constants of other common laser host materials such as YALO, Y_2O_3 , YVO_4 , and YLF could be measured by the same technique that has been employed for YAG.

AFB[®] between dissimilar materials

Since Van der Waals bonds are, contrary to chemical bonds, not localized but are essentially a phenomenon that is based on electro-optical properties such as absorption in the UV and IR, the dielectric constant and the refractive index of a material in contact, the behavior of at least some of the dissimilar composites are explainable. Even a slight thermal expansion coefficient difference between two permanent bonded solid bodies will induce a great amount of stress at the interface if temperature is different from the bond forming temperature. Furthermore, permanent bonds are usually formed at elevated temperature. Cooling the bonded composite to room temperature will inevitably acquire stress. The thermal expansion coefficients of YAG and sapphire are similar but are different in magnitude by at least $2 \times 10^{-6}/^\circ\text{C}$. This difference will induce 20 MPa stress when the temperature is 100°C different from the bonding temperature. The only logical deduction from the apparent stress-free AFB[®] composite being heated to various temperatures is that the stress induced due the differential thermal expansion coefficient is relieved.

We have interferometrically confirmed that dissimilar composites reduce or relieve stress by sliding at the interface during temperature variation. The minimum required conditions for sliding include at least that no permanent bonds are formed at the interface and the sliding surfaces are sufficiently flat and smooth. We have found that after the AFB[®] composites have been heated above T_{critical} , the composite acquires stress of a magnitude proportional to the product of the temperature drop, ΔT , and the difference of the thermal expansion coefficients between the two dissimilar materials, $\Delta\alpha$.

Spontaneous continuous sliding occurs only when two surfaces are ideally flat, smooth, and chemically inert. A real surface even when it is statistically flat (RMS \rightarrow 0) contains microscopic



asperities that distribute in random fashion for isotropic materials and in an anisotropic manner when the materials are anisotropic. We have found that the separation distance, l , between two surfaces is 2.0 Å. That is less than the measured RMS of 2.1 Å for each of the surfaces prior to bonding. It indicates that on a sub-microscopic scale the surface asperities are likely interlocked and elastically strained. Nonetheless, they still slide upon shear in a stick and slide mode.

References

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