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# Ion distribution in pink muscovite: a reply

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## Abstract

Annersten and Hålenius (1976) have presented compelling arguments to show that $Fe^{3+}$ in the Archer's Post muscovite and some other pink muscovites is in octahedral coordination, rather than tetrahedral coordination, as I originally suggested (Richardson, 1975). Their interpretation is reasonable and may indeed prove to be correct; if so, however, it makes explanation of the observed reverse pleochroism in this muscovite difficult.

## Discussion

My analysis of the Archer's Post muscovite (Richardson, 1975) was primarily an attempt to account for its reverse pleochroism, and incidentally, to provide an alternative explanation for pink coloration in some muscovites. My hypothesis, based on an extension of Faye and Hogarth's (1969) study of reverse pleochroic phlogopite and my own spectroscopic work, correlates both color and pleochroism with the presence of tetrahedrally coordinated ferric iron and leads to the conclusion that "*many* pink muscovites should also be reverse pleochroic, and *all* reverse pleochroic muscovites should be pink."

It is not clear, in the absence of paired polarized absorption spectra for either the Sogn or Ultevis muscovite (Annersten and Hålenius, 1976), whether they (like the Archer's Post mica) are both pink and reverse pleochroic, but their qualitative chemical and physical similarities suggest that this may be the case. If so, Annersten and Hålenius' study of those micas and their reinterpretation of my published spectroscopic data lead to a disturbing conclusion.

I will readily agree with their statement that $Mn^{3+}$ contributes to the pink color of all three micas, since it obviously absorbs light preferentially in the short (<600 nm) wavelengths of the visible range and transmits red (>600 nm) light. This disagrees in no way with my own conclusion that $Fe^{3+}$, which absorbs visible light in the same region, also contributes to the pink hues of these and other muscovites. My hypothesis of the primary importance of $Fe^{3+}$ in the pink micas, however, was supported at least in part by the observation that chemical analyses of pink muscovites reported in the mineralogical literature occasionally lack $Mn^{3+}$, but are only rarely iron-free.

It is not the identity of the chromophore which is in question, though, but its structural position, and here it becomes necessary to consider the reverse pleochroism. Annersten and Hålenius suggest that the observed pleochroism in the Archer's Post muscovite is due to variations in the absorption band intensities for $Mn^{3+}$ in distorted octahedral sites with changing orientation. Two arguments against this proposal can be presented:

(1) Examination of the polarized absorption spectra for the Archer's Post specimen (Richardson, 1975; Figure 2) indicates that the differences in absorption intensity between $E\perp(001)$ and $E\|(001)$ spectra (*i.e.*, the pleochroism) are greatest for the absorption bands at $\sim 19600\ cm^{-1}$ and $\sim 22600\ cm^{-1}$. These bands have been identified as $Fe^{3+}$ bands in a number of studies (Faye, 1968; Faye and Hogarth, 1969; Manning, 1972) as well as in Annersten and Hålenius' own study. The absorption bands at $\sim 18000\ cm^{-1}$ and $\sim 13700\ cm^{-1}$, which are $Mn^{3+}$ bands, show much smaller changes in intensity with orientation.

(2) As first proposed by Faye and Hogarth, the mechanism which is currently invoked to explain increased absorption perpendicular to the basal cleavage plane (*i.e.*, "reverse" pleochroism) in trioctahedral micas relies on a vibronic coupling between the $E\perp(001)$ electric vector and the vibrational (stretching mode) state of the $Fe^{3+}_{IV}$–O bond which bridges between the tetrahedral and octahedral sheets. There are no other bonds (other than similar Si–O and $Al_{IV}$–O bonds) parallel to principal optical axes in the mica structure. Unless we accept Faye and Hogarth's

conclusion that such a coupling mechanism enhances $Fe^{3+} \rightarrow O^{2-}$ charge transfer, there is as yet no other way to account for reverse pleochroism. Octahedrally-coordinated transition-metal ions cannot contribute to the effect observed in trioctahedral micas, and in view of structural similarities, it would be surprising if they could contribute to reverse pleochroism in the dioctahedral micas either.

On the basis of these two arguments, it seems unlikely that octahedral $Mn^{3+}$ can account for reverse pleochroism in the Archer's Post mica or in any other pink muscovite. On the contrary, we should expect to find $Fe^{3+}_{IV}$ in reverse-pleochroic dioctahedral micas, in direct analogy to the well-established pattern in trioctahedral micas.

We are faced, therefore, with a dilemma. The spectroscopic results and interpretations presented by Annersten and Hålenius (1976) are quite reasonable. As they point out, the high value for isomer shift, particularly in the light of revised data for Brown and Pritchard's (1969) ferri-orthoclase, strongly suggests octahedral, rather than the tetrahedral coordination which I originally proposed. I also agree that, with somewhat less certainty, the qualitatively low extinction coefficient for the 22624 $cm^{-1}$ band in the optical absorption spectra suggests the presence of octahedral, rather than tetrahedral iron. If it were not for the unusual pleochroism of the Archer's Post sample, I would show no hesitation in accepting their revised interpretation. As it is, either of two equally improbable and diametrically opposed alternatives could be true:

(1) Annersten and Hålenius are correct, and Faye and Hogarth's explanation does not apply to reverse pleochroic dioctahedral micas, or

(2) Faye and Hogarth's mechanism applies to reverse pleochroic muscovite, and Annersten and Hålenius are wrong.

## Conclusion

The interpretation presented by Annersten and Hålenius is consistent with prior studies in the literature of mineral spectroscopy, and raises some serious objections to my own interpretation of optical properties of the Archer's Post muscovite and other similar micas. It leads, however, to fundamental disagreements with the only reasonable explanation for reverse pleochroism proposed to date, without providing a structurally acceptable alternative. In view of this contradiction, it will clearly be necessary to obtain further structural information on the reverse pleochroic muscovites and to reevaluate the mechanism for reverse pleochroism proposed by Faye and Hogarth.

## References

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